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Characterization of Scrubbed and Unscrubbed Power Plant Plumes: Three Case Studies

Meteorology Research, Inc.
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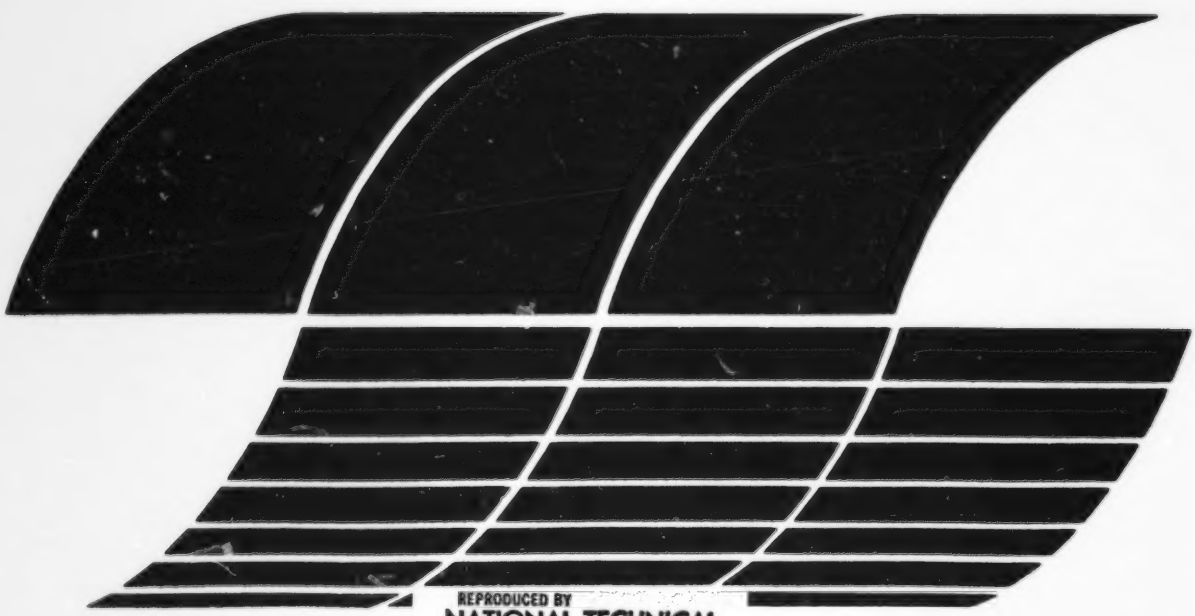
Three Case Studies

Interagency Energy/Environment R&D Program Report



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Three Case Studies

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ABSTRACT

Airborne measurements of scrubbed and unscrubbed emissions from the Widows Creek Power Plant were carried out during August 17-25, 1978. The results of the analysis of the measurements taken during three case study days are described.

Gas-to-particle conversion rates calculated for mixtures of scrubbed and unscrubbed emissions were between 0.3-4% hr^{-1}. Secondary particles were formed in both the nuclei (particles < 0.05 μm) mode in the accumulation mode (0.05 - 1.0 μm). In one instance, in a plume consisting largely of scrubbed emissions in relatively clean background air, most of the secondary particles were produced in the nuclei mode. For the other cases, the amount of aerosol volume formed in the nuclei mode amounted to between 0.2 and 3% of the total secondary aerosol. Day to day variations in ambient sulfate levels were at least as great as the increase in sulfate levels due to the plume. The scrubbed plume was not a significant source of particles greater than 1.0 μm or of primary sulfates in the submicron size range.

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SECTION 1

INTRODUCTION

This report presents the analysis of the results of an airborne sampling program performed by Meteorology Research, Inc. (MRI), as part of a study of scrubbed and unscrubbed power plant plumes (referred to as the SCRUB project).

The production of primary and secondary pollutants has been studied extensively in plumes of unscrubbed power plants, but only preliminary work has been done to determine the effects of scrubbers on downwind plume chemistry. There is mounting political pressure to reduce ambient sulfate levels by reducing the emissions of sulfur dioxide (SO_2), the precursor to secondary (i.e., particles produced by gas-to-particle conversion) sulfate particles, from coal-fired power plants. The primary methods of reducing SO_2 emissions are to use low sulfur coal or to use scrubbers in the stack to reduce the amounts of SO_2 produced. Since scrubbing allows the use of higher sulfur coal, scrubbing is likely to become more common in the future, as utilities utilize the more available higher sulfur coal.

A number of studies have been conducted to determine the rate of gas-to-particle conversion in the atmosphere. A recent review by Hegg (1979) lists the results of 11 separate investigations of the conversion rates of sulfur dioxide; conversion rate estimates in these studies range from 0 - 300% hr^{-1}. However, most recent studies (e.g., Forrest and Newman, 1977; Ursenbach et al., 1977; Cantrell and Whitby, 1978; Gillani et al., 1978; Husar et al., 1978; Pueschell and Van Valin, 1978; Stith, 1978; Hegg, 1979) indicate a conversion rate between 0 and 10% hr^{-1}.

In spite of the many studies of gas-to-particle conversion rates which

have been conducted, there still exists considerable uncertainty as to the formation mechanisms, and the rates of conversion. Husar et al. (1978) and Gillani et al. (1978) have observed sulfate conversion to be directly related to the amount of solar radiation for the coal-fired Labadie Power Plant. Cantrell and Whitby (1978), also at the Labadie plant, found the sulfate fraction to monotonically increase with plume travel time. On the other hand, Forrest and Newman (1977) and Meagher et al. (1978) found very little change in secondary sulfates with plume travel time. Friberg (1978) suggested that conversion proceeds to a fractional asymptotic limit.

Dittenhoffer and Pena (1978) found the production of Aitken particles, in the Keystone coal-fired power plant, to be the dominant secondary production process under conditions of low humidity, near neutral stability, and strong solar radiation; while for plumes of relative humidity near 100%, they found that conversion occurred on pre-existing droplets.

In a study of the Labadie Power Plant, Whitby et al. (1978) found the creation of nuclei mode particles to range from about 1700 $cm^{-3} s^{-1}$ (at 6:30 in the morning) to about 1 $cm^{-3} s^{-1}$ at night. They found that about 5% of the aerosol volume which formed as a result of gas-to-particle conversion went into the nuclei mode, and was independent of the conversion rate. They also found that coagulation of these nuclei with particles in the accumulation (0.05 - 1.0μm) mode reduced the mean size of particles in the accumulation mode in the plume as compared to the background air.

One of the main mechanisms for the oxidation of SO_2 (as well as NO_2) is oxidation by OH radicals. The rate of oxidation by this mechanism is between 0.4-2.7% hr^{-1} for SO_2 (Calvert et al, 1978) and about 13% hr^{-1} for NO_2 (Cox, 1974). Since the OH radicals are formed photochemically, the rate of conversion should depend on the amount of solar radiation.

SO_2 oxidation may also take place on the surface of existing particles (e.g., carbon particles) or in water droplets. This liquid phase oxidation may play an important role under cloudy or high humidity conditions (Hegg

and Hobbs, 1978). Further, metal salts in fly ash act as catalysts for the oxidation of SO_2 dissolved in water droplets. Thus, the SO_2 conversion rates may be affected by any droplets produced by the scrubber.

The aerosol formation mechanisms in scrubbed plumes may be different from the unscrubbed plumes as a result of the different primary emissions. The objective of the present study is to determine what differences, if any, exist in the plume chemistry, aerosol size distributions, aerosol formation rates, and transformations of aerosols among scrubbed, unscrubbed, and mixed plumes.

SECTION 2

CONCLUSIONS

The principal findings that have emerged from this study are:

- Particles are formed by gas-to-particle conversion in both the nuclei and accumulation modes. In one case, where the background air was comparatively clean, the most significant increase in particulate volume, in a plume which most likely consisted primarily of scrubbed emissions, occurred in the nuclei mode. In another case, with similar primary emissions but with very dirty ambient air, most of the increase occurred in the accumulation mode. For the latter case, the secondary particulate volume in the nuclei mode was about 0.3-0.6% of the total secondary particulate volume.
- In a mixture of scrubbed emissions and fly ash laden emissions (owing to problems with the electrostatic precipitators), the fraction of secondary particulate volume in the nuclei mode reached a maximum of ~3% at 24-25 Km downwind and then decreased to ~0.2% at 75 Km.
- The differences between the scrubbed and unscrubbed emissions, aside from the expected lower SO_2 concentrations, were: the scrubbed emissions probably contained fewer primary particles greater than 0.05 μm in size than did the combined emissions; and the NO_2/NO ratio may be higher in the scrubbed plume. The differences, in

terms of the size of the secondary aerosols which are produced, are probably relatively minor compared to the day to day variations.

- The rates of gas-to-particle conversion of SO_2 which were observed in both types of plumes is similar to that reported for other power plants (between 0 and 4% hr^{-1}).
- Day to day variations in ambient sulfate levels for this particular region were at least as great as the increase in sulfate levels caused by the plume, for both types of plumes.
- The scrubbed plume is not a significant source of primary sulfates in the submicron size region.
- The scrubbed plume is not a significant source of coarse ($> 1.0\ \mu m$) particles.
- Significant ozone formation downwind of the Widows Creek plant was observed. Since above ambient ozone was detected in both a plume which contained a large scrubbed fraction and in a mixed plume, the formation of ozone may be more influenced by ambient conditions than by the differences between the scrubbed and unscrubbed primary emissions.
- Regions of high condensation nuclei concentration were observed in the Widows Creek plume. One of them coincided with a region of relatively high NO_2 concentrations.

SECTION 3

BACKGROUND: THE 1978 SCRUB PROGRAM

THE WIDOWS CREEK POWER PLANT

The power plant studied in the SCRUB project was the Tennessee Valley Authority's (TVA) Widows Creek steam plant near Stevenson, Alabama. The plant has six 135 MW units (units 1-6) which are connected to a single 305 m stack. Two additional 557 MW units are each connected to separate 152 m stacks. One of these units (#8) is equipped with a limestone wet scrubber, with a design efficiency of 80% for SO_2 removal. Units 1-6 and unit #8 are equipped with electrostatic precipitators with design efficiencies of greater than 99.5%. The precipitators on unit 7 have a design efficiency of 90%.

DATA COLLECTION: INSTRUMENTATION AND METHODOLOGY.

The aircraft used in this study was the MRI Beechcraft Queen Air, which contains instrumentation for measuring atmospheric trace gases and aerosols, as well as several meteorological parameters. A list of the instruments aboard the Queen Air is presented in Table 1. Location of some of the instruments on the aircraft are shown in Figure 1.

Data from all continuous instruments, as well as time and event codes, are recorded on 9-track tape in digital form. These tapes are then processed at MRI where the appropriate calibration factors are applied to the data. Further, altitude corrections are made to correct for changes in the zero and span response of various instruments as a function of altitude.

TABLE 1. QUEEN AIR INSTRUMENTATION

Parameter	Sampler Manufacturer and Model	Analysis Technique	Normal Measurement Ranges (Full Scale)	Time Response (to 90%)	Approximate Resolution
SO_2	Meloy 285	Flame Photometric	50, 100 ppb	30 s	1 ppb
NO/NO_x *	Monitor Labs 8440	Chemiluminescence	200, 500 ppb	5-10 s	< 10 ppb
O_3	CSI	Chemiluminescence	200 ppb	5 s	5 ppb
Sulfate	MRI Two-Mass	Flash vaporization/ flame photometric	> 3 and < 3 μm dia		~ 1 $\mu g\ m^{-3}$
"	ERT RSP Sampler	Ion Chromatograph	< 3 μm dia		~ 1 $\mu g\ m^{-3}$
Light Scattering Coefficient	MRI 1550	Integrating Nephelometer	$10^{-6}\ m^{-1}$	1 s	$0.1 \times 10^{-4}\ m^{-1}$
"	MRI 1569	"	10^{-4} and $10^{-3}\ m^{-1}$ (Dual Range)	< 1 s	$10^{-6}\ m^{-1}$
Condensation Nuclei	Environment One Rich 100	Light Attenuation	$10^5\ cm^{-3}$	3 s	$10^3\ cm^{-3}$
Turbulence	MRI 1120	Pressure Fluctuations	0-10 $cm^{2/3} s^{-1}$	3 s (to 60%)	0.1 $cm^{2/3} s^{-1}$
Temperature	YSI/MRI	Bead Thermistor/ Vortex Housing	-50° to +50° C	5 s	0.5° C

* Not installed while the Meloy 285 Particulate Sulfur Monitor was in use.

TABLE 1 (continued)

Parameter	Sampler Manufacturer and Model	Analysis Technique	Normal Measurement Ranges (Full Scale)	Time Response (to 90%)	Approximate Resolution
Dew Point	Cambridge Systems 137	Cooled Mirror	-50° to +50° C	0.5 s/°C	0.5° C
Altitude	Validyne	Absolute Pressure Transducer	0-3000 m msl	1 s	6 m
Indicated Airspeed	Validyne	Differential Pressure Transducer	23-68 m s^{-1}	1 s	0.1 ms^{-1}
Position	King KX170B/ HTI DVOR	Aircraft DME/VOR	0 to 359° and 0 to 150 km from the station	1 s	1° (bearing), 0.2 km (distance)
Data Logger (includes time)	MRI Data System	9-Track Tape - 6 hour capacity in continuous operation	0 to ±9.99 V DC		0.01 VDC
Stripchart Recorder	Linear Instruments	Dual channel	0.01, 0.1, 1, 10 VDC	< 1 s	
Aerosol Charge Acceptance	Washington University	Aerosol Charge Acceptance	~ .01-.1μ particles	~ 1 s	
Particulate Sulfur*	Meloy 285	Upstream SO_2 Scrubber/Measurement of total sulfur	66 $\mu g\ S\ m^{-3}$	30 s	Relative changes in particulate sulfur only

* Not installed while the Monitor Labs 8440 NO/NO_x Monitor was in use.

TABLE 1 (continued)

AEROSOL SIZE DISTRIBUTION MEASUREMENTS		
Instrument	Size Range	Method
TSI 3030*	.0056 - 1 μm	Aerosol Charger/Mobility Analysis
Royco 218/MCA*	.56 - 18 μm	Optical Particle Counter with Multichannel Analyzer
Knollenberg ASSP	3 - 45 μm	Axial Scattering Spectrometer Probe
Lundgren Impactor (modified by the University of Calif. at Davis)	10 to < 1/2 μm	4 stage impactor with final filter. Diameters of 50% cutoff: 4, 2, 1, and 0.5 μm.

* Automatic bag sampling system for TSI 3030 and Royco 218; bagfill requires about 4 seconds

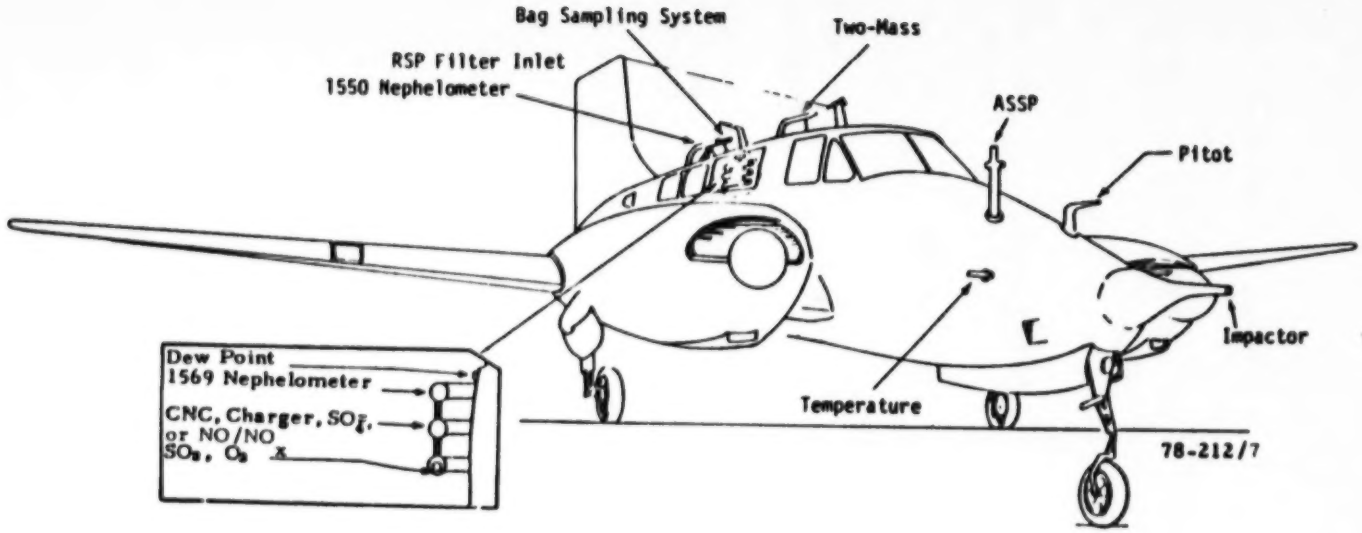

Figure 1. MRI Queen Air sample inlet lines and external probes.

Three types of flight maneuvers were used in this study: traverses, spirals, and orbits. Traverses are used to determine the horizontal distribution of the various measured parameters, while spirals are used to determine their vertical distribution. Generally, when sampling a plume, such as the plume from the Widows Creek plant, these traverses are done perpendicular to the axis of the plume, at various altitudes and distances downwind. In this way, the three-dimensional structure of the plume can be determined for cases where the plume does not change radically during the period of the sampling mission. The spirals done upwind of the plant are especially useful for determining background concentrations and the structure of the mixing layer. Orbits are done to provide filter or impactor samples at one specific location.

Ground reference points are used to determine the endpoints of each traverse, and the location of each spiral.

For producing plume cross sections, the data from the traverses are plotted in vertical or horizontal cross sections, using the endpoints of each traverse to fix the location of the aircraft. A contour analysis is then done. Since the data for one cross section requires ~15-60 minutes to collect, these cross sections represent the true plume cross sections only for steady state plumes.

Particulate sulfur (sulfate) concentrations were determined by flash volitization and flame photometric analysis of Two-Mass filters and by ion chromatography analysis of RSP filters. The filter material for the Two-Mass samples is glass fiber, the RSP filters are Teflon coated quartz. When activated, the filter samplers sample continuously from the sampling ports outside the aircraft (Figure 1). The flow rates are about 50 and 25 l m^{-1} for the RSP and Two-Mass samplers, respectively. All sample volumes were reduced to standard conditions of 25° C and 1013.2 mbar. Both samplers have a similar particle size cutoff (~3 μm). A comparison of RSP data with the Two-Mass data, collected at the same time, is presented in Figure 2. These data are from a combination of in-plume and background samples.

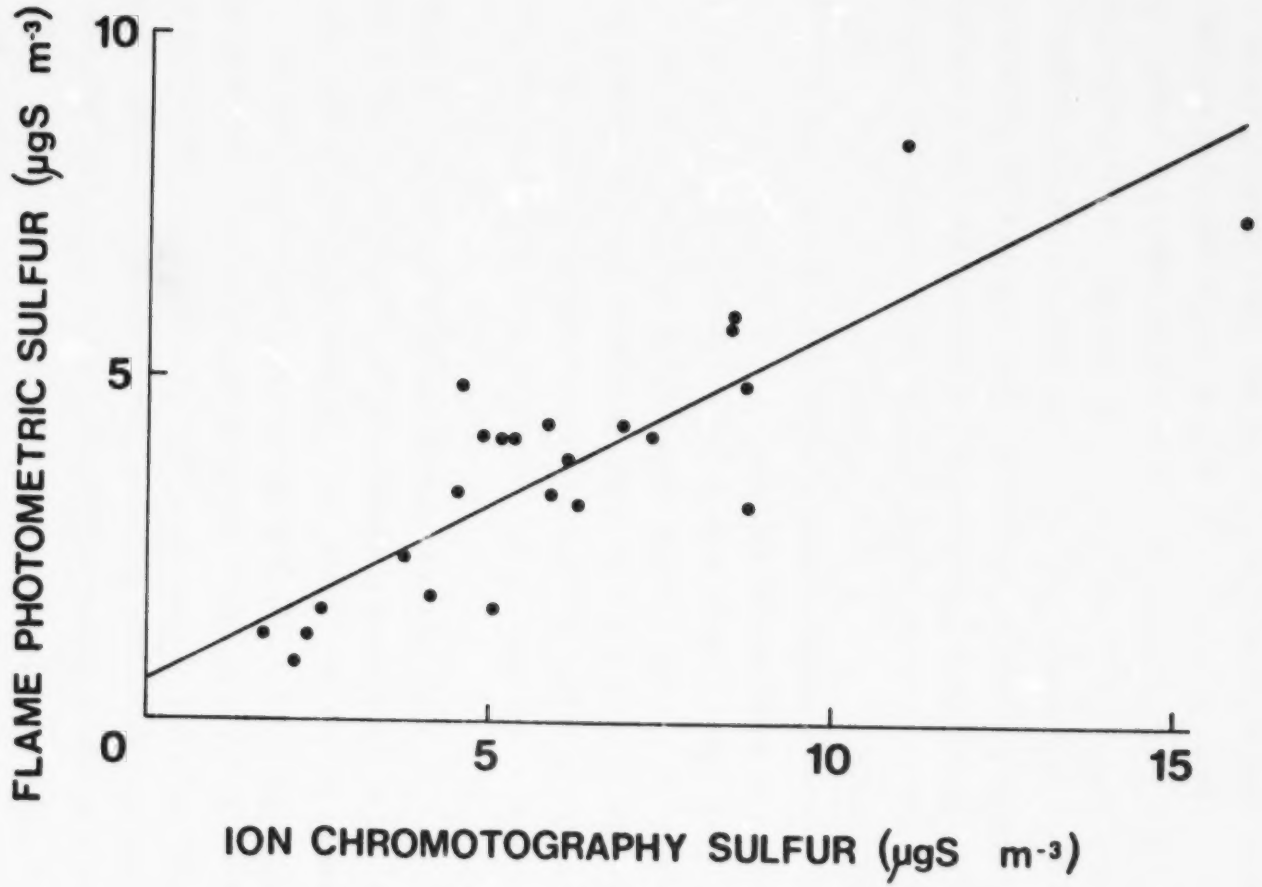

Figure 2. Comparison of the sulfate concentrations from ion chromatography analysis of RSP filters versus the sulfate concentrations determined by flash volitization/flame photometric analysis of Two Mass filters. Each filter was exposed over the same period of time. The slope of the regression line which fits the data is 0.52; the correlation coefficient is 0.86.

The two methods are reasonably correlated (correlation coefficient of 0.86); however, the sulfur concentrations derived from the RSP filters are, on the average, about twice as high as the Two-Mass results. Since many more Two-Mass filters were generally exposed than were RSP filters, sulfate concentrations used in this study are from the Two-Mass results.

RSP filters were also analyzed for nitrate by ion chromatography. These results are somewhat uncertain, as the nitrate levels on the filters are near the values found on blank filters.

Elemental analyses of the Lundgren impactor samples were also conducted using Ion Excited X-ray Emission (IEXE). Some of these results are available in the SCRUB data volume (Stith, et al., 1979); additional results are available upon request to MRI. Sulfur concentrations determined by the IEXE analyses were often higher than RSP or Two-Mass values.

The systematic difference between the Two-Mass, RSP, and impactor results have been consistent through several field programs. At present, the ion chromatography analyses of RSP filters appear to give the most reasonable results; however, these differences have not yet been resolved.

SO_2 conversion rates were calculated following the procedure used by Husar et al. (1978): the conversion rate is given as the charge in the fraction of total sulfur present as sulfate per unit plume travel time. This method will overestimate the conversion rate if there is significant removal of SO_2 before the measurements are made. Most of the plumes sampled in this study were either above the terrain entirely, or had the main portion of the plume above the terrain; consequently, little removal of SO_2 should have taken place. These conversion rates may also be low (perhaps by as much as a factor of 2), since the sulfate concentrations which are used are from the Two-Mass results. Plume travel times were determined from the average windspeed at plume elevation and the distances downwind. Windspeeds were determined by pibal measurements made by TVA. More details on these calculations are provided in Appendix A.

Particle size distributions are presented as number concentration in each instrument channel (dN), normalized by the finite difference between the base 10 log of the instrumental channel boundaries (d log D), versus the geometric mean of the channel boundaries. Particle surface and volume distributions are computed from the number distributions, assuming spherical particles.

Following Whitby (1978), we refer to three modes which describe the aerosol size distribution. These modes appear as three additive log-normal functions, and are most easily observed on the particle surface or volume distributions, with the nuclei mode in the 0.005 - 0.05 μm range, the accumulation mode in the 0.05 - 1.0 μm range, and the coarse particle mode greater than 1.0 μm. Nuclei mode particles are usually fresh condensation products from chemical or high temperature (e.g., combustion) processes, while accumulation mode particles result from condensation and from coagulation of nuclei mode particles. Coarse particles are produced usually by grinding or impaction or may result from windblown dust.

Particles formed by photooxidation of SO_2 are usually in the nuclei mode range, unless there is sufficient aerosol surface area (greater than $\sim 500\ \mu m^2\ cm^{-3}$) that condensation on existing particles, rather than the formation of new particles, is the dominant particle formation mechanism. For more details see, for example, Whitby (1978) or Bouland et al. (1978).

Gas-to-particle conversion by liquid phase oxidation should produce particles in the accumulation mode size range, owing in part to the original size of the droplet condensation nucleus, which for most natural clouds is in the range 0.05 - 0.5 μm.

SUMMARY OF SAMPLING PROGRAM

A total of ten flights were conducted by MRI at the Widows Creek site during August 17-25, 1978. A short summary of the flights made by MRI is given in Table 2. The data collected by MRI are available in the form of a Data Volume (Stith et al., 1979) and on magnetic tape.

TABLE 2. SCRUB PROGRAM SUMMARY

Cart #/ Date	Flight Objective	Sampling Period (CDT)	Plume Sampling Distances (km)	No. of Traverses/ Spirals/ Orbits*	No. of Samples			Comments
					RSP	Impactor	Two-Mass	
29/ 17 Aug	Characterization of unit #8 (scrubbed) plume	0845-1224	0.5, 3, 9 downwind	0/2/16	3	5	8	Unit #7 not operating
30/ 17 Aug	Duplication of AM sampling for AM/ PM comparison	1551-191	0.5, 2, 7	0/2/15	4	5	7	Unit #7 not operating
31/ 19 Aug	Long range plume characterization	0630-1108	0.5, 9, 20, 45, 75	10/3/10	6	5	13	Good plume characterization; Precipitators out of service.
32/ 19 Aug	Continuation of AM flight	1407-1741	1, 9, 13, 17, 25, 30	8/3/7	2	2	10	Vertical structure determination at ~25 km downwind; near source characterization; plume spread out and patchy beyond 10 km; Precipitators out of service.
33/ 21 Aug	NO_y plume chemistry determination; comparison flights with TVA helicopter	0647-1031	15-20, 30	14/2/1	3	3	7	Plume spread out and shifting - poor characterization data
34/ 22 Aug	Sunrise experiment on unit #8 plume	0609-0835	0.5, 10	6/2/10	1	1	4	Experiment terminated due to low clouds and fog; limited sampling within 10 km of plant; poor plume definition - shifting winds
35/ 23 Aug	Long range plume characterization flight; sampling in conjunction with TVA	0915-1322	15, 50	13/2/2	5	5	6	Good plume characterization data; documentation of NO, NO_x and O_3 chemistry; documentation of plume O_3 bulge; unit #7 not operating

*Circles around the stacks are included as orbits.

TABLE 2 (continued)

Cart #/ Date	Flight Objective	Sampling Period (CDT)	Plume Sampling Distances (km)	No. of Traverses/ Spirals/ Orbits*	No. of Samples			Comments
					RSP	Impactor	Two-Mass	
36/ 23 Aug	Continuation of AM sampling; sampling in conjunction with TVA	1441-1907	13, 50, 70	7/1/5	1	1	13	Unit #7 not operating
37/ 25 Aug	Sunrise study - document history of plume and background before, during, and after sunrise	0511-0936	1, 8, 20	2/2/19	0	7	8	Sun first visible 0620; close in plume samples were of the plumes from units #7 and #8
38/ 25 Aug	Continuation of AM flight	1030-1316	43-50, 70	6/1/4	0	4	4	Good (light) plume definition at 48 km not found at 70 km

*Circles around the stack are included as orbits.

Other groups have participated in the SCRUB program. TVA performed airborne sampling of trace gases and particles using an instrumented helicopter. The University of Minnesota performed similar sampling from an instrumented van. TVA also provided plant operation and meteorological data. Midwest Research, Inc. performed emission characterization measurements on the scrubbed unit.

Several of the flights conducted by MRI provide detailed case studies of the scrubbed and unscrubbed plumes. On August 17 and 23 morning and afternoon flights were made. On these days unit #7 (the largest unscrubbed unit) was not operating. The data from these days provide case studies of scrubbed and mixed plumes. On August 19 , morning and afternoon flights were made while all the units were operating; however, some of the precipitators were out of service These primary emissions should be quite different from the August 17 and 23 emissions.

In the following chapters, the results of the analyses of these case studies are presented.

SECTION 4

CASE STUDY: AUGUST 17, 1978

BACKGROUND

On August 17, 1978, morning and afternoon flights were conducted while the largest unscrubbed unit (#7) was not operating. Thus, much of the sampling was in the scrubbed plume.

The weather in the area was characterized by warm humid flow over the area accompanied by afternoon and evening convective activity. A radiation inversion was present in the area during the early morning hours, breaking up about 0930 CDT. Ground fog was present in the area prior to this time. The afternoon was unstable with light precipitation from late afternoon thundershowers. Winds during the morning near plume elevation were between 2 - 6 m s^{-1} from the west-southwest. The wind died down during the afternoon.

Aircraft sampling was conducted near the plant by flying circles around the stacks, penetrating the plume at about 0.5 km downwind. In-plume orbits were then conducted at a near (3 km) range and a far (9 km during the morning flight and 7 km during the afternoon flight) range. Shallow background spirals, covering the plume vertical extent were done to provide data in background air.

The plume from the tall stack (units 1 - 6, unscrubbed) was observed to be above the aircraft during the passes at 0.5 Km. At the further distances, the tall stack plume may have mixed with the lower plume.

The plume during the morning flight was relatively well defined; however, during the afternoon the plume was more spread out. Traces

of old plume may have also been in the area.

During the period 0500 - 2400 (CDT), unit #8 (scrubbed) was operating at a relatively steady level with hourly gross generation between 275 - 320 MW.

PARTICLE SIZE DISTRIBUTIONS

Many bagfills were collected during the plume orbits, and the particle size distributions determined. Tables 3 and 4 summarize these results in terms of the particle volume concentration in the nuclei and accumulation modes, along with the sulfate concentrations which were measured while these bagfills were obtained. Examples of the particle number, surface, and volume distributions are presented in Figure 3, for the morning flight. As is clearly evident from Table 3 and Figure 3, there was a significant increase in the nuclei mode concentration at the 9 km distance during the morning flight. At the 3 km distance, the nuclei mode concentrations were similar to that in the background. Accumulation mode concentrations were little different from background levels at both ranges, for the morning flight. A coarse particle mode (particles greater than 1.0 μm) is also seen in both the plume and background (Figure 3). The coarse mode is highly variable, hence the slight differences in this mode in Figure 3 are not a result of the plume.

The results from the afternoon flight were quite different from the results obtained during the morning. Although, on average, there was a slight increase in the nuclei mode, the differences are not significant as were the earlier results. SO_2 concentrations were about a factor of 2 lower than at similar distances downwind during the morning.

The nuclei mode concentrations were generally lower in the afternoon, both in the plume and out, than they were in the morning. The accumulation mode was greater in the afternoon than in the morning. These results indicate that the afternoon airmass had characteristics of a more aged airmass than did the airmass present during the morning. It is difficult,

TABLE 3. SUMMARY OF SIZE DISTRIBUTION MEASUREMENTS
TAKEN ON THE MORNING OF AUGUST 17, 1978

Time	Nuclei Mode Volume Concentration ($\mu m^3 cm^{-3}$)	Accumulation Mode Volume Concentration ($\mu m^3 cm^{-3}$)	SO_2 Concentration (ppb)	Sulfate * Concentration ($\mu gS\ m^{-3}$)	Comments
10:24:39	0.16	10.	20.		
10:31:01	0.13	8.9	41.	0.89	
10:39:00	0.087	6.9	62.		3 km downwind
11:01:07	0.095	6.0	30.		457 m altitude
11:05:24	0.14	6.4	41.	1.3	Plume orbit
11:08:58	0.16	6.7	51.		
11:11:57	0.098	6.6	45.		
	0.12 , 0.031	7.4 , 1.5	41. , 14.	1.1 , 0.29	Average and standard deviation at 3 km
11:28:52	0.64	8.6	57.		
11:34:17	0.69	11.	112.	1.5	9 km downwind
11:40:11	0.52	8.8	42.		457 m altitude
11:44:31	0.46	6.5	29.	1.2	Plume orbit
11:53:57	0.56	11.	24.		
	0.57 , 0.092	9.2 , 1.9	53. , 35	1.4 , 0.21	Average and standard deviation at 9 km
09:51:22	0.083	9.8			
09:54:27	0.048	7.4	~0.5	0.75	Background
09:57:10	0.089	5.3			426 - 495 m
	0.073 , 0.022	7.5 , 2.3	~0.5	0.75	Average and standard deviation of background samples

* Sulfate concentrations are from Two-Mass filters which were being exposed while the bag samples were obtained; often several bagfills were obtained while one filter was being exposed.

TABLE 4. SUMMARY OF SIZE DISTRIBUTION MEASUREMENTS TAKEN ON THE AFTERNOON OF AUGUST 17, 1978

Time	Nuclei Mode Volume Concentration ($\mu m^3\ cm^{-3}$)	Accumulation Mode Volume Concentration ($\mu m^3\ cm^{-3}$)	SO_2 Concentration (ppb)	Sulfate * Concentration ($\mu gS\ m^{-3}$)	Comments
17:32:43	0.070	14.	23.		
17:40:32	0.069	12.	17.	1.6	3 km downwind 457 m altitude Plume orbit
17:45:34	0.029	14.	40.		
17:51:48	0.078	12.	16.	1.4	
	0.062 , 0.022	13. , 1.2	24. , 11.	1.5 , 0.14	Average and standard deviation at 3 km
18:29:03	0.046	11.	30.		
18:33:40	0.080	13.	41.	4.9	
18:37:07	0.050	8.8	17.		7 km downwind 457 m altitude Plume orbit. Possible portion of another plume.
18:41:03	0.086	11.	13.	1.6	
18:44:55	0.090	13.	50.		
18:48:16	0.046	9.2	14.		
	0.066 , 0.021	11. , 1.8	28. , 15.	3.3 , 2.3	Average and standard deviation at 7 km
16:56:26	0.033	9.8			
16:59:03	0.013	9.8	~0.5	1.3	Background sampling 400 - 500 m
17:01:50	0.073	10.			
17:05:09	0.078	8.2			
	0.049 , 0.031	9.4 , 0.84	~0.5	1.3	Average and standard deviation of background samples

* Sulfate concentrations are from Two-Mass filters which were being exposed while the bag samples were obtained; often several bagfills were obtained while one filter was being exposed.

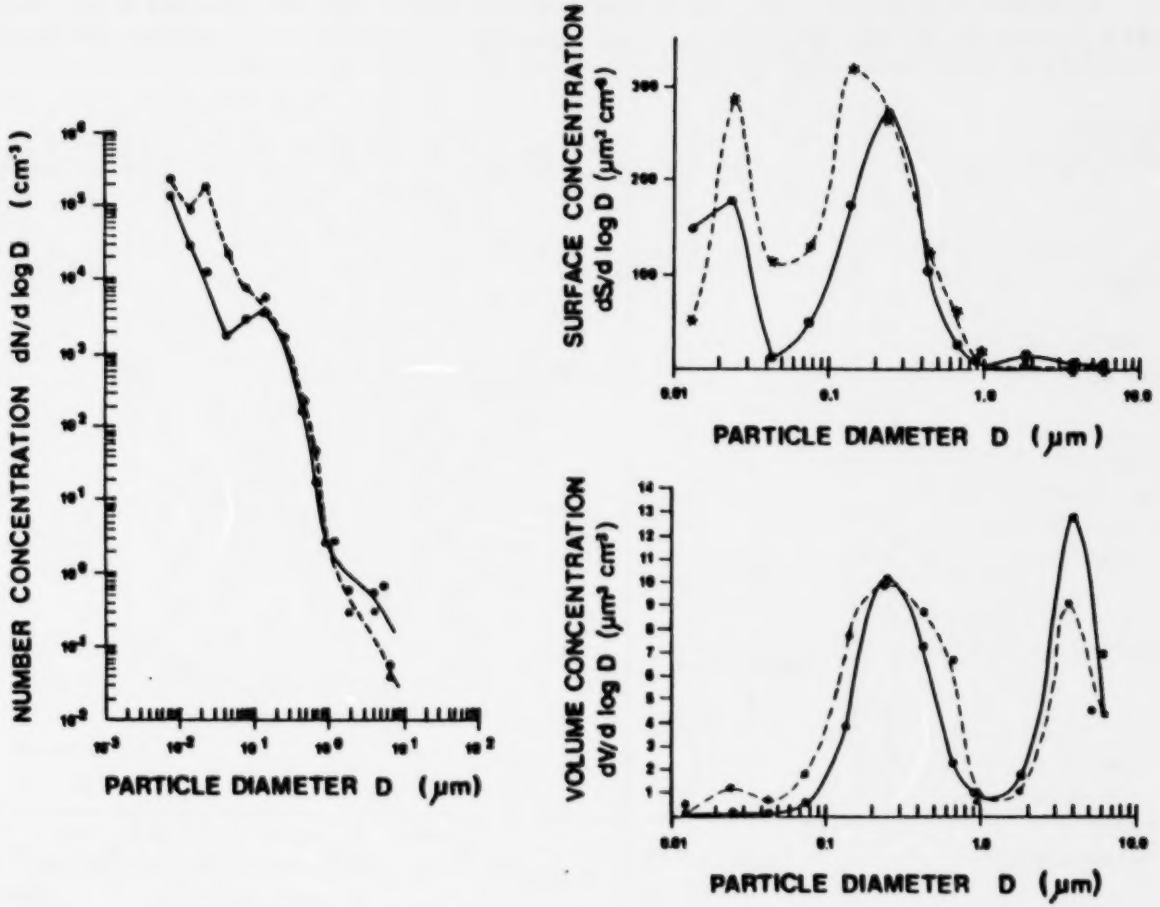
NUMBER CONCENTRATION dN/d log D (cm⁻³)
PARTICLE DIAMETER D (μm)
SURFACE CONCENTRATION dS/d log D (μm² cm⁻³)
PARTICLE DIAMETER D (μm)
VOLUME CONCENTRATION dV/d log D (μm³ cm⁻³)
PARTICLE DIAMETER D (μm)

Figure 3. Particle number (dN/dlog D), surface (dS/dlog D), and volume (dV/dlog D) distributions, measured in scrubbed emissions from the Widows Creek Power Plant on August 17, 1978 at 9 km downwind (dotted line) and at an altitude of 457 m (MSL). A distribution taken in the background air at an altitude of 459 m is also presented (solid line).

however, to compare the in-plume results obtained in the afternoon to the morning results, since the afternoon plume was more spotty, and may have mixed with traces of old plume (of some sort) which was in the area.

GAS-TO-PARTICLE CONVERSION

Sulfate levels in-plume during the morning flight generally increased to about twice the background level (Table 3). Two sulfate filters were exposed during each plume orbit during the morning flight. The sulfate concentrations measured during the 3 Km orbit were 0.89 and 1.3 $\mu gS\ m^{-3}$. Above ambient total sulfur concentrations measured while these filters were exposed were 39 and 54 $\mu gS\ m^{-3}$, respectively. The ambient sulfate concentration was 0.75 $\mu gS\ m^{-3}$. Thus, the fraction of total sulfur as sulfate at the 3 Km distance was 0.36 and 1.0%, or an average of 0.68% with standard deviation of 0.45%. At the 9 Km distance the sulfate concentrations were 1.5 and 1.2 $\mu gS\ m^{-3}$. Above ambient total sulfur concentrations measured while these filters were exposed were 63 and 28 $\mu gS\ m^{-3}$, respectively. Thus, the fraction of total sulfur as sulfate at the 9 Km distance was 1.2 and 1.6%, or an average of 1.4% with standard deviation of 0.28%. Based on a windspeed of 3 m s^{-1}, which existed during the sampling period, this results in a gas-to-particle conversion rate of $1.3 \pm 0.73\%\ hr^{-1}$. However, these results may be low by a factor of 2, as a result of the systematic errors discussed in Section 3. A discussion of the uncertainty in this value is given in Appendix A.

Background sulfate levels were about a factor of 2 higher in the afternoon than during the morning flight. It is difficult to estimate a conversion rate from these data, since the sulfate concentrations in the plume were generally not very different from background values (Table 4). Further, the winds died down during the afternoon.

SECTION 5

CASE STUDY: AUGUST 19, 1978

BACKGROUND

On August 19, 1978, morning and afternoon flights were conducted while all of the units were operating. However, only about half the precipitators on units 1-6 were functioning, and the precipitator on unit 7 was shut down. Consequently, this plume presents a much more particulate laden plume than was present earlier.

A maritime tropical airmass was present in the area, associated with a subtropical high pressure system over the southeastern United States. A radiation inversion was developed to about 650 meters altitude, during the morning hours, with final breakup occurring by about 1000 CDT. Winds at plume elevation were between 2 - 4 m s^{-1}, out of the northwest during the morning. Windspeed was slightly higher during the afternoon and was out of the southwest. No precipitation occurred.

During the morning flight, aircraft sampling near the plant was conducted by flying circles around the stack and penetrating the plume at about 0.5 km downwind. The aircraft then moved out to successively further distances downwind, where a series of traverses through the plume and orbits in the plume were conducted. The plume was followed out to a distance of about 75 km. In all, plume sampling was done at 0.5, 9, 8, 24, 25, 45, 47, 70, and 75 km downwind. Thus, the morning flight provides a very good long range characterization of the plume.

During the afternoon another long range characterization flight was attempted. However, the wind had shifted, and the plume was much more

broken up and hard to follow. Consequently, the sampling which was done was rather limited. The results presented in this section are from the morning flight.

Units 7 (unscrubbed) and 8 (scrubbed) were operating at a relatively steady load during the morning flight, with operating levels between 305 - 330 MW and 245 - 250 MW for units 7 and 8, respectively. During the period 0900 - 1000 the output of units 1 - 6 (unscrubbed, tall stacks) increased from about 450 to 690 MW. In-stack opacity on unit #7 increased considerably during the period 0600 - 0800, which was due to the shutdown of the precipitators.

PLUME CROSS SECTIONS

Cross sections of the concentrations of SO_2, O_3, and condensation nuclei (CN) and the light scattering coefficient due to particles, b_{sp}, measured at 24 km downwind, are presented in Figures 4 to 7, respectively. It is evident from these cross sections that the lower portion of the plume was much different than the upper portion. The lower region contained less SO_2; hence, it may likely be the plume from the scrubbed stack. However, both regions of the plume had a similar impact on the b_{sp}. The possibility of contamination from some other source of CN cannot be ruled out; however, it is highly unlikely because the CN plume and the plume from the power plant exactly coincide. At the 75 km distance downwind, a slight ozone bulge (~ 10 ppb) was observed.

Another unusual aspect of the plume is that the lower portion of the plume contained higher levels of NO_2 relative to NO than did the rest of the plume. These results are summarized in Table 5. As can be seen in Table 5, this ratio is consistently higher in the lower portion of the plume. The usual increase in this ratio with distance downwind is also noted, a result of conversion of the NO produced by the plant into NO_2 by ambient ozone. The observed ozone deficit in the plume is approximately equal to the amount of NO which has been converted to NO_2. Virtually all of the

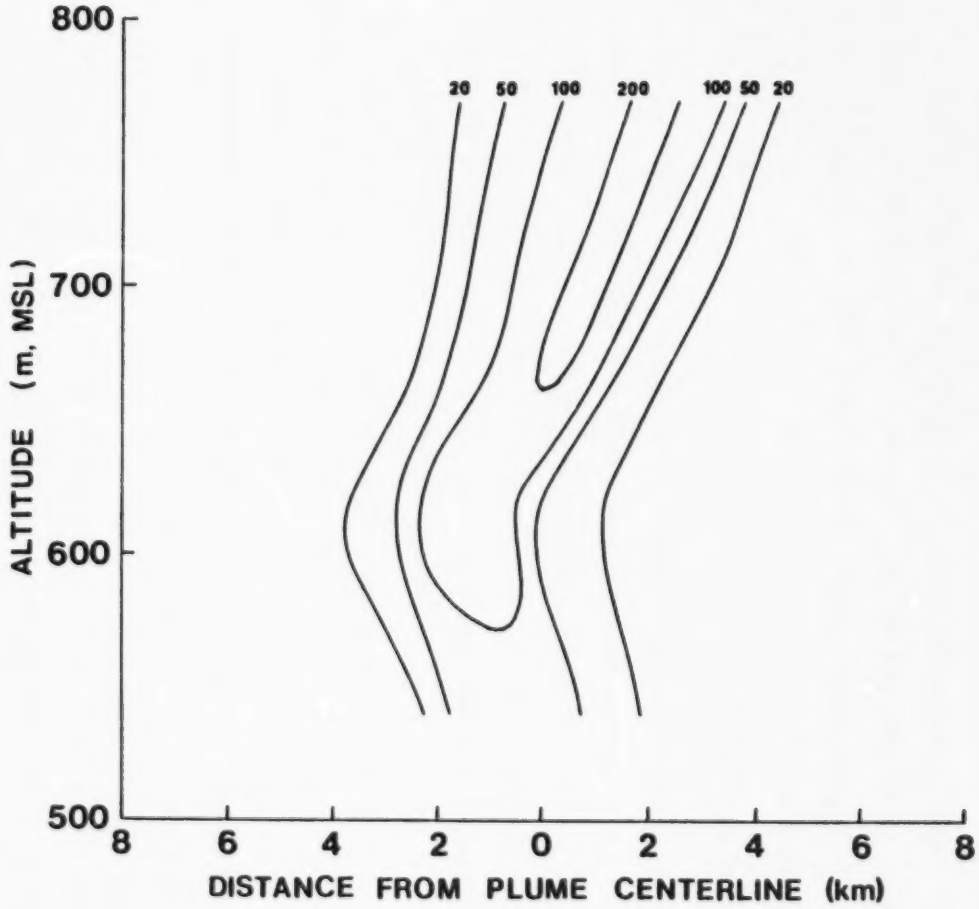

Figure 4. Contours of sulfur dioxide concentration (in ppb) measured perpendicular to the axis of the plume from the Widows Creek Power Plant on August 19, 1978 at 24 km downwind. The plume was comprised of a mixture of scrubbed and unscrubbed emissions. The concentration of sulfur dioxide in background air was between 0 and 5 ppb.

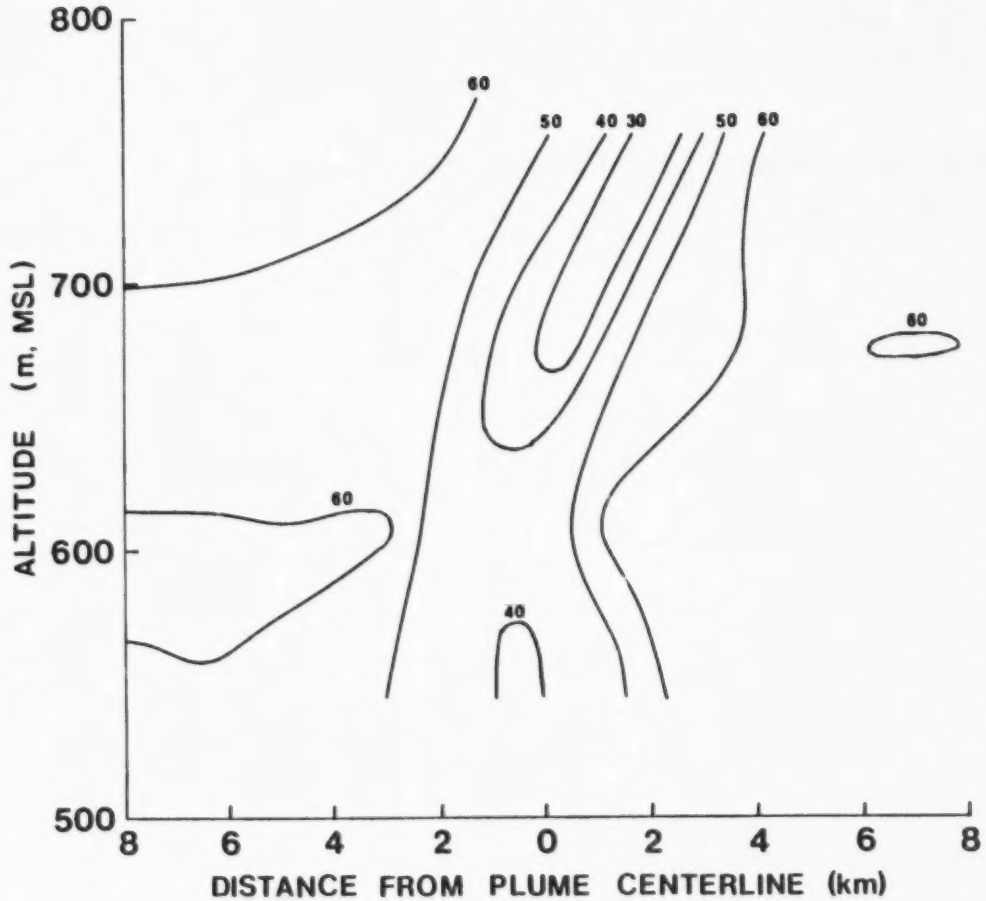

Figure 5. Contours of ozone concentration (in ppb) measured perpendicular to the axis of the plume from the Widows Creek Power Plant on August 19, 1978 at 24 km downwind. The plume was comprised of a mixture of scrubbed and unscrubbed emissions. The ozone concentration in background air was between 56 and 70 ppb.

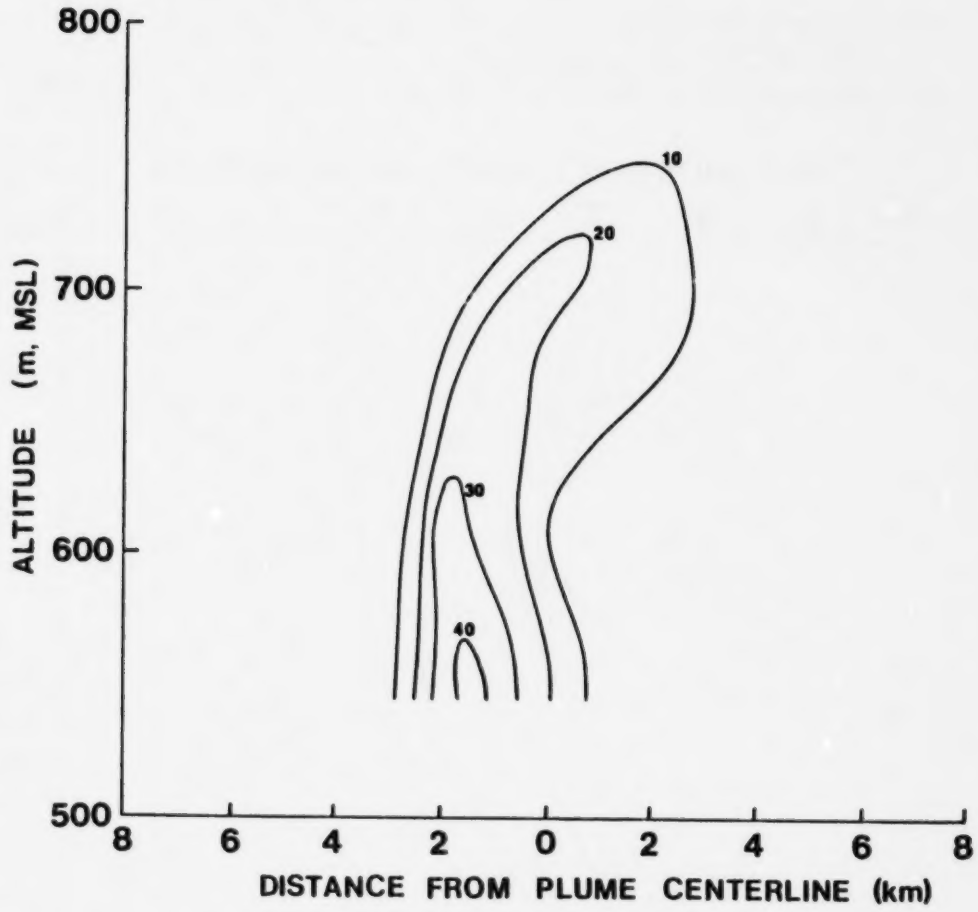

Figure 6. Contours of condensation nuclei concentration (in units of 10^3 cm^{-3}) measured perpendicular to the axis of the plume from the Widows Creek Power Plant on August 19, 1978 at 24 km downwind. The plume was comprised of a mixture of scrubbed and unscrubbed emissions. The background CN concentration was between 3×10^3 and 10^4 cm^{-3}.

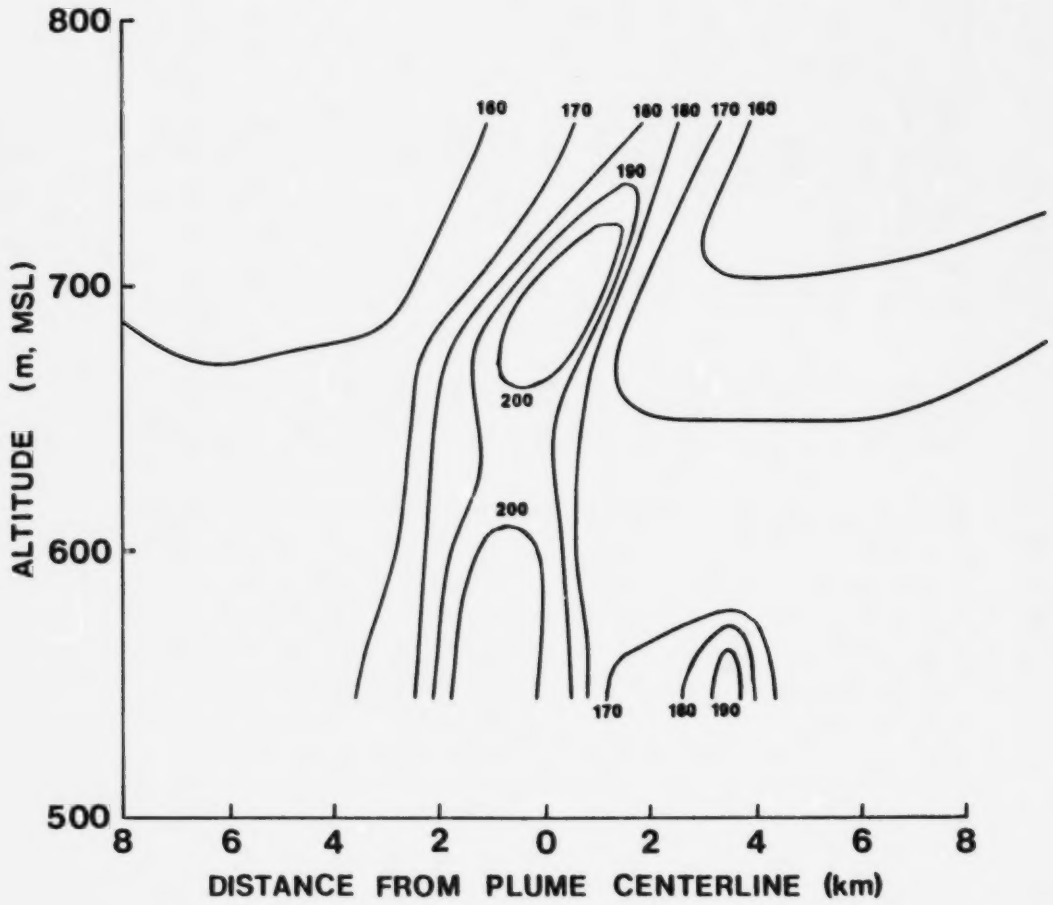

Figure 7. Contours of light scattering coefficient due to particles (in units of 10^{-6} m^{-1}) measured perpendicular to the axis of the plume from the Widows Creek Power Plant on August 19, 1978 at 24 km downwind. The plume was comprised of a mixture of scrubbed and unscrubbed emissions.

TABLE 5. SUMMARY OF NO_2/NO RATIOS IN THE WIDOWS CREEK POWER PLANT ON AUGUST 19, 1978

Altitude (m, MSL)	Distance Downwind/ Travel Time km/hr.	Peak NO_2/NO Ratio	SO_2 Peak Concentration (ppb)
762	~0.5/0.05	0.24	428
670	~0.5/0.05	0.19	1377
610	~0.5/0.05	0.20	1398
549	~0.5/0.05	0.32	1393
488	~0.5/0.05	1.78	292
610	9/0.8	0.32	703
549	9/0.8	0.33	1387
488	9/0.8	1.62	333
762	24/2.2	1.45	221
671	24/2.2	1.60	206
610	24/2.2	2.08	149
549	24/2.2	4.33	114

NO_2 which was present in the upper portions of the plume at the 24 km distance could be accounted for as having resulted from this conversion. However, for the lower portion of the plume (549 m, altitude), there was an excess (~14 ppb at 24 km downwind) of NO_2. This indicates that one of the units may have been a primary source of NO_2. This may result from electrical discharges occurring in a precipitator which is not functioning properly. The electrical discharge produces ozone which quickly reacts with NO to increase the NO_2 level. If the observed NO_2 excess is due to precipitator problems, the unit which is responsible is probably the scrubbed unit, since the precipitators on unit 7 (unscrubbed) were shut down and the excess occurred in the region of lower SO_2 concentration near the bottom of the plume. It is interesting to note that this region of higher NO_2 coincides with the region of high CN concentrations.

PARTICLE SIZE DISTRIBUTIONS

A summary of some of the results of the size distribution analyses of bagfills which were obtained at the various distances downwind at roughly similar altitudes are presented in Table 6. Sulfate concentrations which were measured while the bagfills were being taken are also presented.

As is seen in Table 6, the nuclei mode concentration remained relatively constant until 75 km, where the concentration decreased to near background levels. In contrast to the August 17 results, the accumulation mode concentrations were significantly higher than background levels. The accumulation mode concentrations remained relatively constant out to the 75 km range. However, since the plume was diluting as it moved out to the 75 km range, it is clear that there was creation of particulate volume in both the nuclei and accumulation mode ranges by gas-to-particle conversion. Using (from Table 6) the average plume above ambient nuclei and accumulation mode concentrations, and normalizing by the average plume above ambient SO_2 concentration, we see that this ratio was 1.2×10^{-4}, 0.0013, 0.0032, and 4.2×10^{-4} μm^3 cm^{-3} ppb^{-1} for the nuclei mode and 0.028, 0.044, 0.11, and 0.26 μm^3 cm^{-3} ppb^{-1} for the accumulation mode at the 9, 24-25, 45, and 75

TABLE 6. SUMMARY OF SIZE DISTRIBUTION MEASUREMENTS TAKEN ON THE MORNING OF AUGUST 19, 1978

Time	Nuclei Mode Volume Concentration ($\mu m^3 cm^{-3}$)	Accumulation Mode Volume Concentration ($\mu m^3 cm^{-3}$)	SO_2 Concentration (ppb)	Sulfate * Concentration ($\mu gS\ m^{-3}$)	Comments
08:21:06	0.27	56.	1388.	7.4	9 km downwind, 610 m altitude
08:25:03	0.070	25.	639.		
	0.17 , 0.14	40. , 22.	1014. , 530.	7.4	Average and standard deviation at 9 km
08:55:01	0.087	19.	197.	2.8	24 - 25 km downwind, 671 m altitude
09:16:59	0.39	17.	108.		
09:21:57	0.21	19.	138.	3.7	
09:26:54	0.25	18.	105.		
	0.23 , 0.12	18. , 0.96	137. , 43.	3.25 , 0.64	Average and standard deviation at 24 - 25 km
10:05:15	0.10	19.	76.	3.9	45 km downwind, 610 m altitude
10:18:30	0.24	19.	67.	4.9	
10:23:28	0.51	22.	76.		
	0.28 , 0.21	20. , 1.7	73. , 5.2	4.4 , 0.71	Average and standard deviation at 45 km

* Sulfate concentrations are from Two-Mass filters which were being exposed while the bag samples were obtained; often several bagfills were obtained while one filter was being exposed.

TABLE 6 (continued)

Time	Nuclei Mode Volume Concentration ($\mu m^3 cm^{-3}$)	Accumulation Mode Volume Concentration ($\mu m^3 cm^{-3}$)	SO_2 Concentration (ppb)	Sulfate * Concentration ($\mu gS\ m^{-3}$)	Comments
10:42:36	0.091	19.	27.		
10:45:20	0.074	18.	26.	4.2	75 km downwind 610 m altitude
10:48:03	0.042	16.	25.		
10:50:46	0.038	23.	28.		
	0.061 , 0.026	19. , 2.9	27. , 1.3	4.2	Average and standard deviation at 75 km
06:37:05	0.038	14.			
06:40:23	0.054	13.	~0.5	2.6	Background 439 - 879 m
06:58:49	0.059	10.		2.4	
	0.050 , 0.011	12. , 2.1	~0.5	2.5 , 0.14	Average and standard deviation of background samples

* Sulfate concentrations are from Two-Mass filters which were being exposed while the bag samples were obtained; often several bagfills were obtained while one filter was being exposed.

km distances downwind, respectively. Thus, though the volume in the nuclei mode was initially increasing at a faster rate than the volume in the accumulation mode, most of the volume was created into the accumulation mode. The fraction of secondary particulate volume in the nuclei mode was about 0.4, 3, 3, and 0.2% at the 9, 24-25, 45, and 75 km distances, respectively.

These results are similar to what is typically observed in smog chambers (e.g., Whitby et al., 1978; Bouland et al., 1978): the concentration of nuclei tends to increase until the point where the formation rate is equal to the coagulation rate, and then the concentration decreases. Further, in particularly dirty air (where the particle surface area is greater than or equal to ~500 μm^2 cm^{-3}), the secondary products may condense directly on the surface of existing particles rather than form new particles. Since the surface area concentration on August 19 was generally between 100 - 500 μm^2 cm^{-3} in the plume, much of the secondary products may have formed on existing accumulation mode particles (most of the surface area was in the accumulation mode, as in Figures 8 and 9).

Examples of some of the particle number, surface, and volume distributions are presented in Figures 8 and 9 for the 24 and 75 km distances downwind. A background size distribution is also included in Figure 9 Unlike the earlier scrubbed plume samples from August 17, a significant coarse particle mode is evident at the 24 km range (Figure 8), which was most likely a result of the shutdown of the precipitators. This was not observed at the 75 km range, however. Unit 7 should normally be a source of coarse particles, since it has a precipitator of only 90% efficiency. The generally higher particle concentrations in the accumulation mode in both the plume and the background air as compared to August 17 are also evident in Figures 8 and 9.

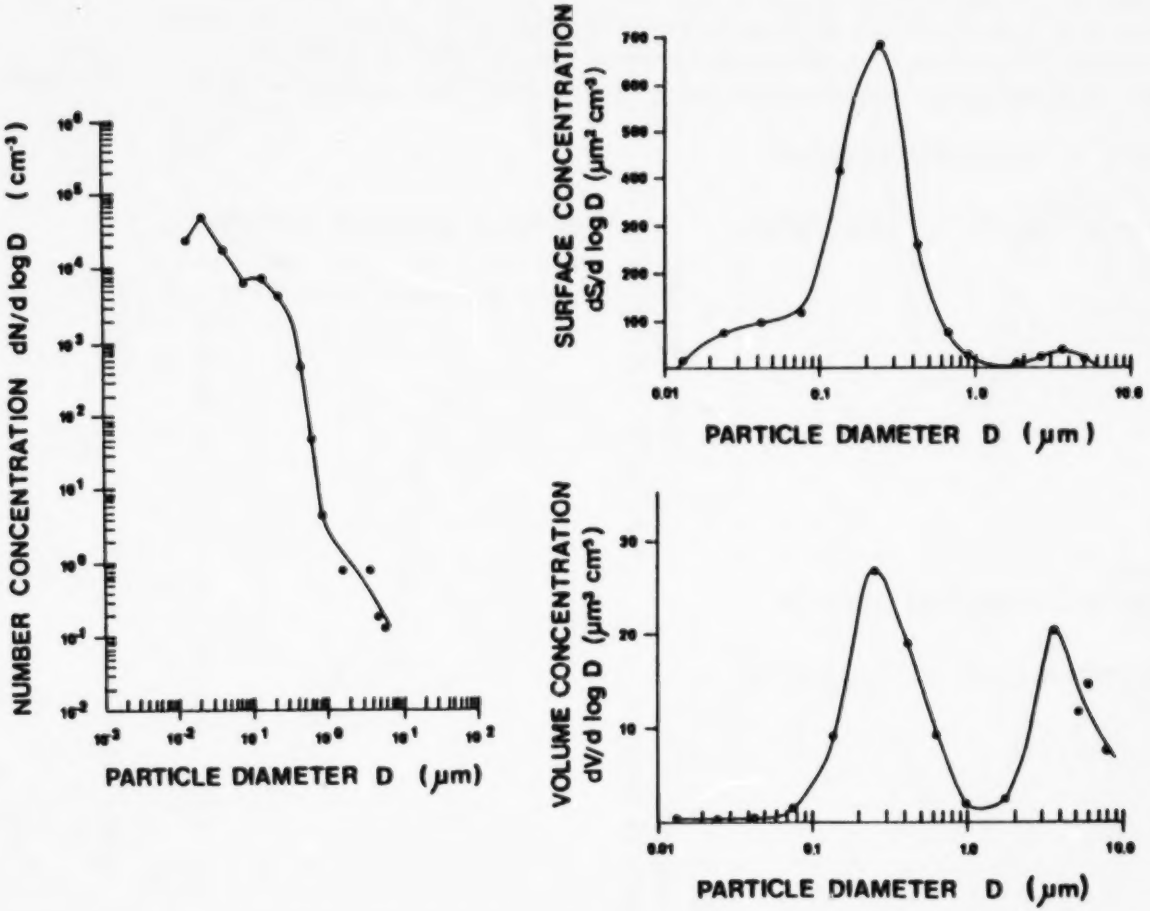
NUMBER CONCENTRATION dN/d log D (cm⁻³)
PARTICLE DIAMETER D (μm)
SURFACE CONCENTRATION dS/d log D (μm² cm⁻³)
PARTICLE DIAMETER D (μm)
VOLUME CONCENTRATION dV/d log D (μm³ cm⁻³)
PARTICLE DIAMETER D (μm)

Figure 8. Particle number (dN/dlog D), surface (dS/dlog D), and volume (dV/dlog D) distributions, measured in a mixture of scrubbed and unscrubbed emissions from the Widows Creek Power Plant on August 19, 1978 at a distance of 24 km downwind and an altitude of 671 m (MSL).

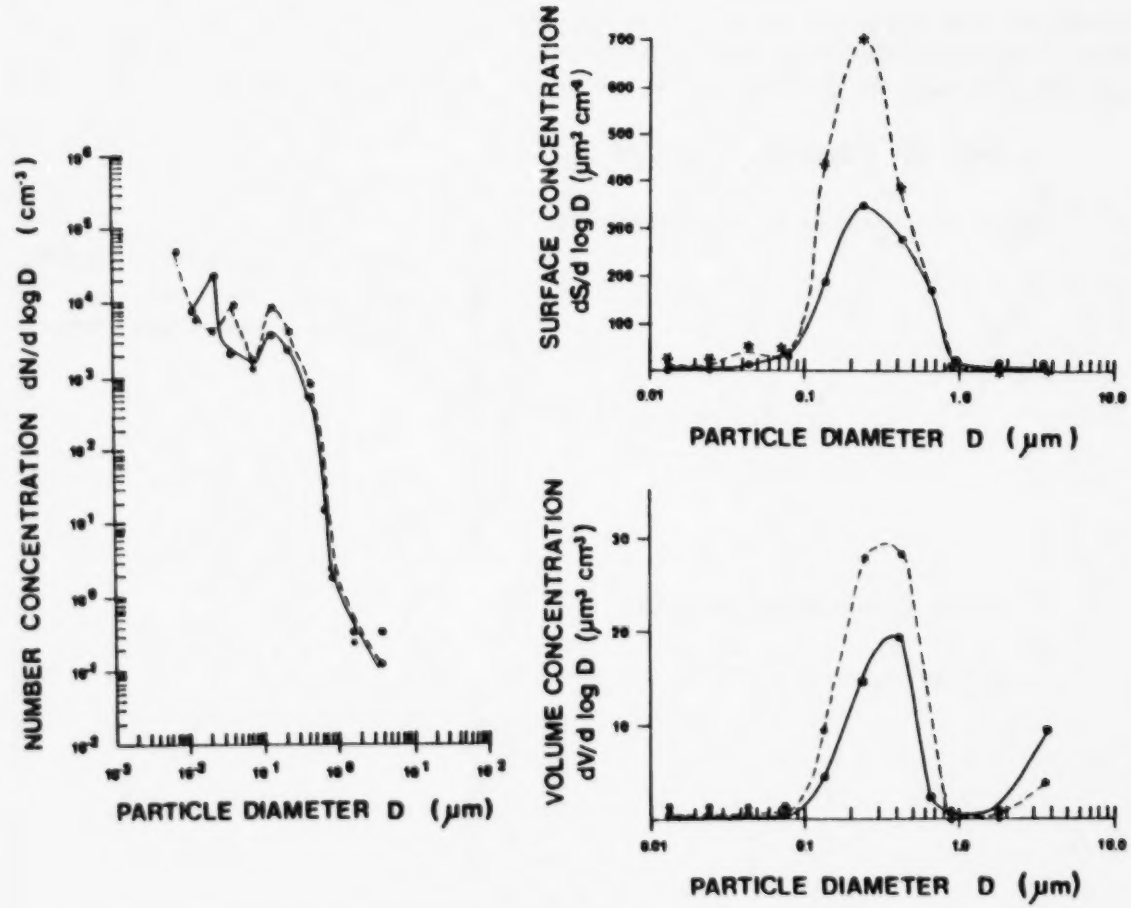
NUMBER CONCENTRATION dN/d log D (cm⁻³)
PARTICLE DIAMETER D (μm)
SURFACE CONCENTRATION dS/d log D (μm² cm⁻³)
PARTICLE DIAMETER D (μm)
VOLUME CONCENTRATION dV/d log D (μm³ cm⁻³)
PARTICLE DIAMETER D (μm)

Figure 9. Particle number (dN/dlog D), surface (dS/dlog D), and volume (dV/dlog D) distributions, measured in a mixture of scrubbed and unscrubbed emissions from the Widows Creek Power Plant on August 19, 1978 at a distance of 75 km downwind and an altitude of 610 m (MSL) (dotted line). A distribution taken in the background air at an altitude of 422 m is also included (solid line).

GAS-TO-PARTICLE CONVERSION

Sulfate levels both in the plume and in the background air were much higher than on August 17. The best data for plume chemistry were obtained during orbits which were done in the background air and in-plume at 8, 25, 45, and 75 km downwind. The sulfate concentrations which were measured during the orbits are 7.4, 3.7, 4.9, and 4.2 $\mu gS\ m^{-3}$ and the corresponding above ambient total sulfur concentrations are 1230, 147, 92, and 36 $\mu gS\ m^{-3}$ at the 8, 25, 45, and 75 km distances, respectively. The background sulfate concentration at plume elevation was 2.4 $\mu gS\ m^{-3}$. Thus, the fraction of above ambient sulfur as sulfate steadily increased downwind; this ratio was 0.41, 0.88, 2.7, and 5.0% at the 8, 25, 45, and 75 km distances, respectively. Based on the average windspeed of 3 m s^{-1} this gives conversion rates of: 0.3% hr^{-1} between 8 and 25 km, 0.99% hr^{-1} between 25 and 45 km, and 0.83% hr^{-1} between 45 and 75 km. The average conversion rate is about 0.71% hr^{-1} with a standard deviation of 0.36% hr^{-1}. The standard deviation provides a measure of the errors inherent in the determination of the conversion rate and of the natural variation in this rate which may result from differences in the amount of solar radiation received by the plume (these emissions were produced between about 0400 to 1100 CDT). Further, these results may be low by about a factor of 2, as a result of systematic errors in the sulfate measurements, as discussed in Section 3.

SECTION 6

CASE STUDY: AUGUST 23, 1978

BACKGROUND

As was the case during the earlier flights on August 17, the main unscrubbed unit (#7) was not operating on August 23. Thus, the sampling which was done was in either the scrubbed plume or the mixed plume. Morning and afternoon flights were made.

The background air was quite polluted with levels of b_{sp} between 2.5×10^{-4} to 3.5×10^{-4} m^{-1}. The radiation inversion which had formed by early morning to about 800 meters had broken up entirely by about 0930. Winds at plume elevation were rather strong ($\sim 6 - 7$ m s^{-1}) during the early morning hours, but by 1000 had died down to about 3 m s^{-1} from the southerly direction where they remained for the rest of the sampling period. Scattered to broken clouds were in the area, with bases at about 1700 m; no precipitation was noted.

During the morning flight a series of traverses were made at 11 - 14 km and at 50 km downwind of the plant. A spiral through the plume and a background orbit were also done. During the afternoon flight a series of traverses and orbits were made starting at 70 km downwind, and these were then repeated at 50 and 13 km downwind.

During the period of these measurements the scrubbed unit (#8) was operating at a steady level (235 - 245 MW, hourly gross generation). Units 1 - 6 (tall stack, unscrubbed) were also operating at a steady hourly gross generation level of about 775 MW.

PLUME CROSS SECTIONS

Cross sections of the concentrations of SO_2, O_3, CN, and b_{sp} for the morning flight are presented in Figures 10 to 13 for the 11 km distance downwind, and in Figures 14 to 17 for the 50 km distance. These cross sections are drawn perpendicular to the axis of the plume looking toward the south (i.e., facing the power plant).

The lower levels of SO_2 in this plume (Figure 10) compared to the levels measured on August 19 (Figure 4), at over twice the distance downwind, are evident. This is probably a consequence of the shutdown of unit #7. The small plume to the side of the main plume is probably the plume from a nearby pulp mill, rather than a portion of the Widows Creek plume. The SO_2 concentrations in the plume on August 23 at 11 km downwind were higher than were measured in the August 17 plume at 9 km downwind, even though on both days unit 7 was shutdown. Evidently the scrubbed plume on August 23 had mixed with the plume from units 1-6, while the emissions sampled on August 17 may have been entirely from the scrubbed unit. Different mixing with the background air may also be a factor. At 11 km range, the Widows Creek plume only had a rather slight effect on the levels of b_{sp} (Figure 13, while at the 50 km distance it had a much greater impact (Figure 17).

As can be seen from the cross sections of ozone concentration (Figures 11 and 15), the ozone levels in the plume went from a deficit to a slight increase between the 11 and 50 km downwind distance. The concentration of CN in the plume at the 50 km distance was about the same as the levels at the 11 km distance (Figures 12 and 16), even though the plume had diluted considerably. Clearly the formation of new CN was occurring by gas-to-particle conversion.

Cross sections of the afternoon plume, looking down from above, are presented in Figures 18 to 21. The plume created an ozone excess of considerable extent downwind of the plant (Figure 19). Generally, b_{sp}

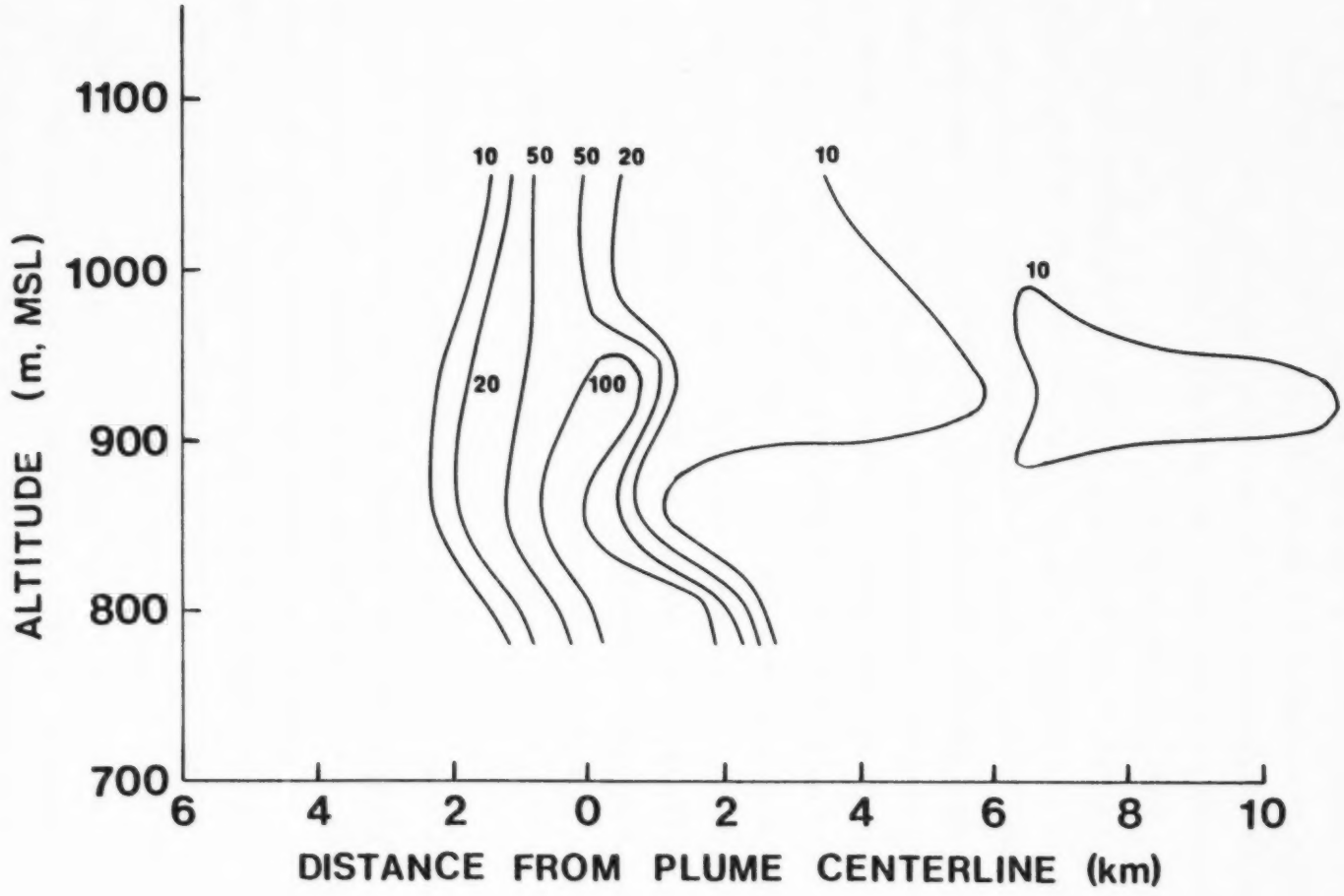

Figure 10. Contours of sulfur dioxide concentration (in ppb) measured perpendicular to the axis of the plume from the Widows Creek Power Plant on August 23, 1978 at 11 km downwind. The concentration of sulfur dioxide in background air was between 0 and 5 ppb.

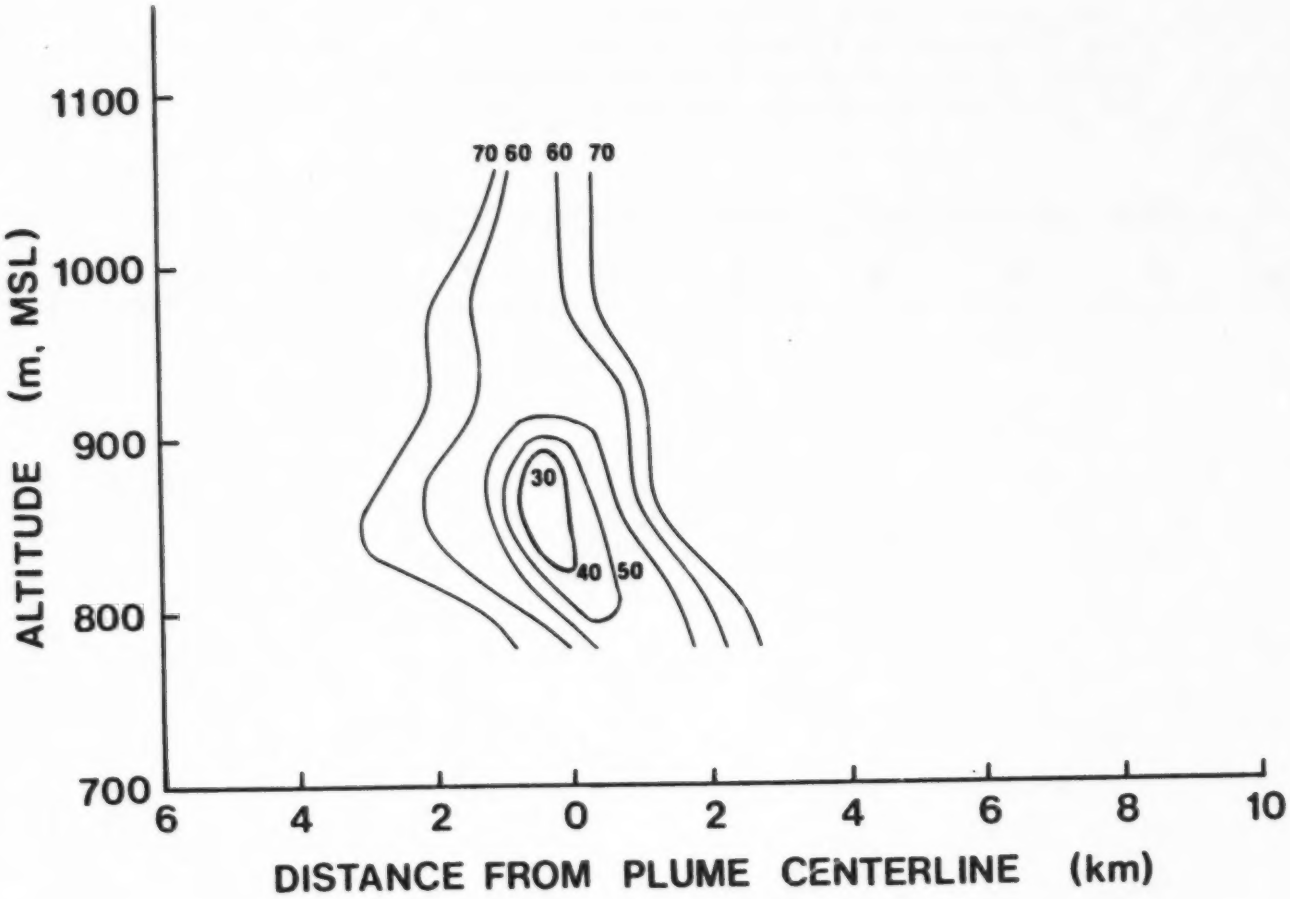

Figure 11. Contours of ozone concentration (in ppb) measured perpendicular to the axis of the plume from the Widows Creek Power Plant on August 23, 1978 at 11 km downwind. The ozone concentration in background air was between 70 and 85 ppb.

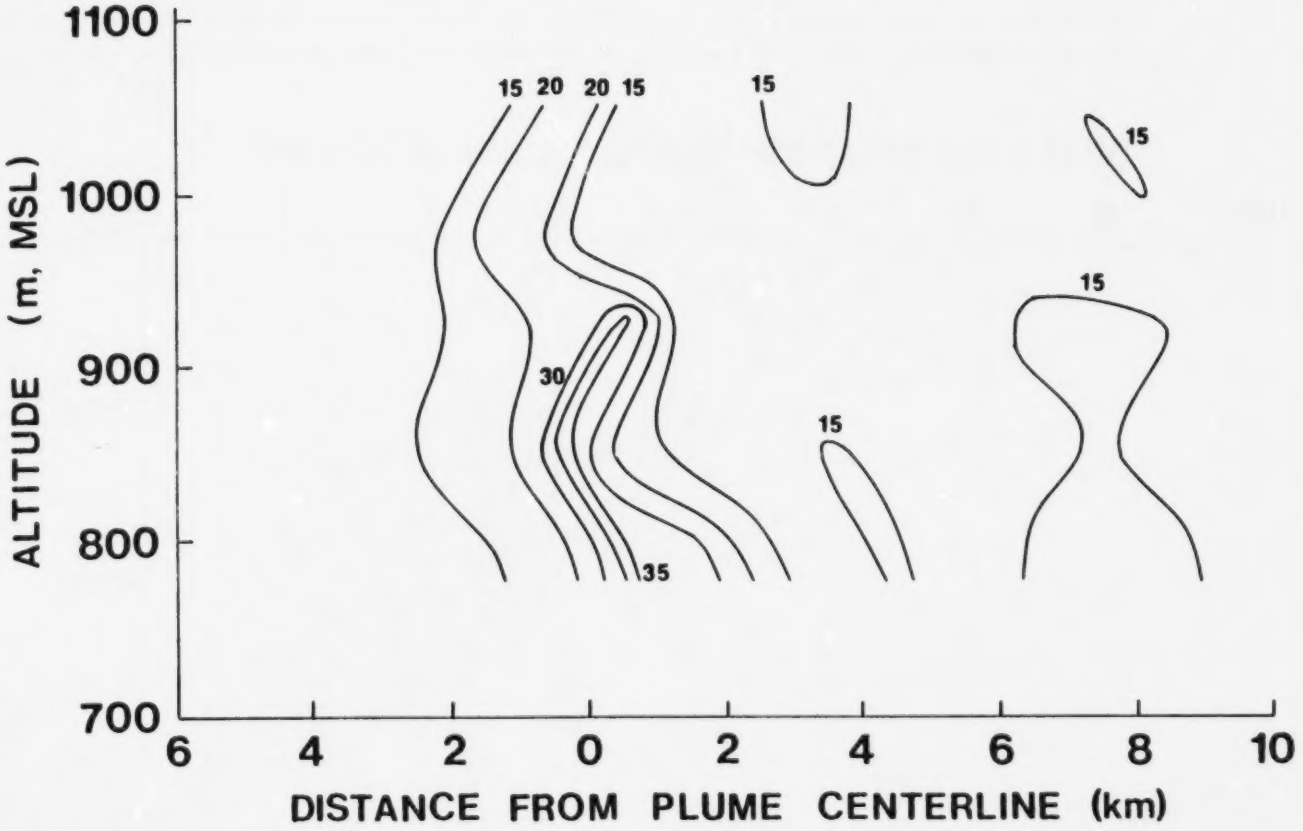

Figure 12. Contours of condensation nuclei concentration (in units of 10^3 cm^{-3}) measured perpendicular to the axis of the plume from the Widows Creek Power Plant on August 23, 1978 at 11 km downwind. The concentration of CN in background air was between 8×10^3 and 15×10^3 cm^{-3}.

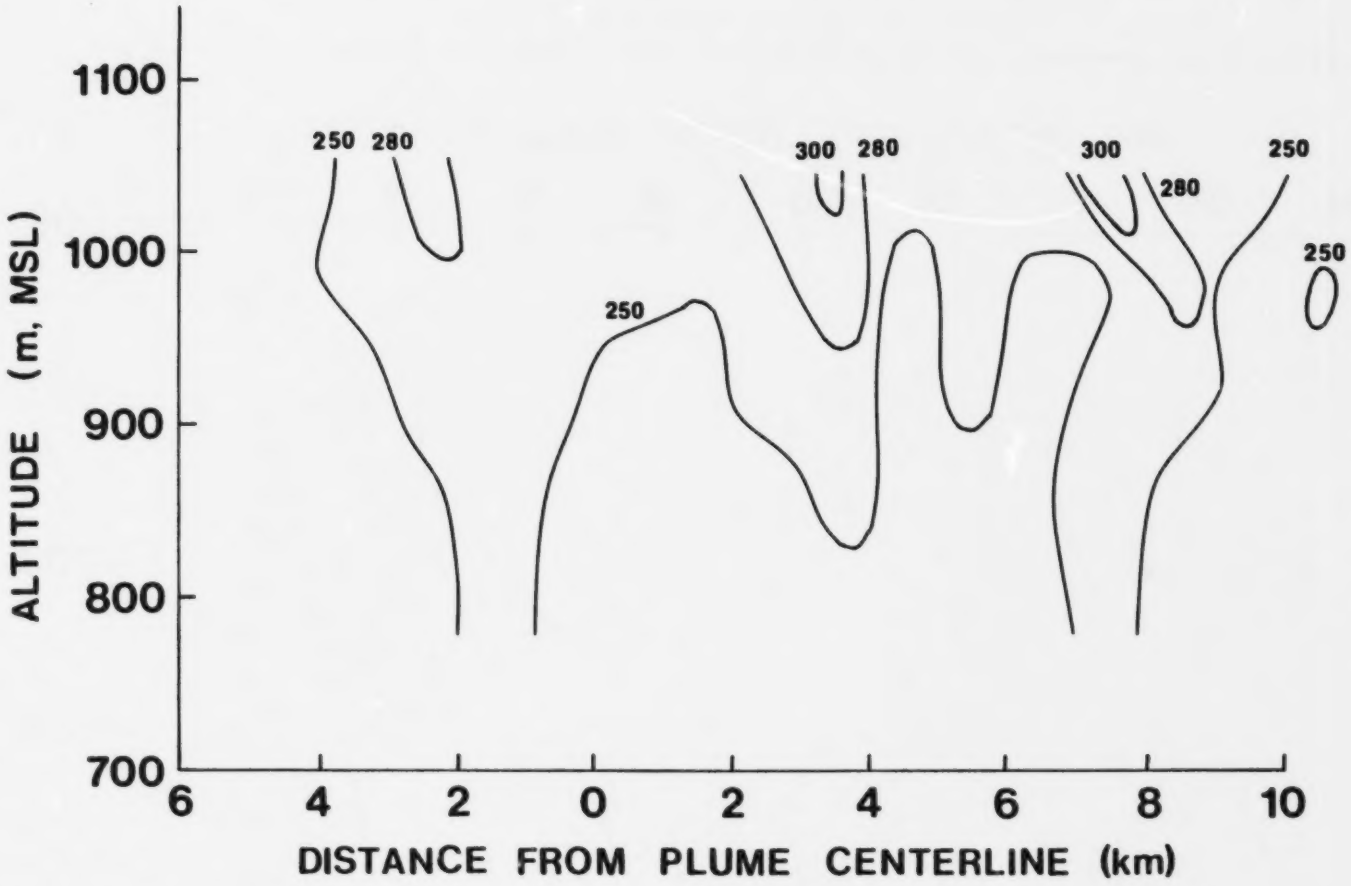

Figure 13. Contours of light scattering coefficient due to particles (in units of 10^{-6} m^{-1}) measured perpendicular to the axis of the plume from the Widows Creek Power Plant on August 23, 1978 at 11 km downwind.

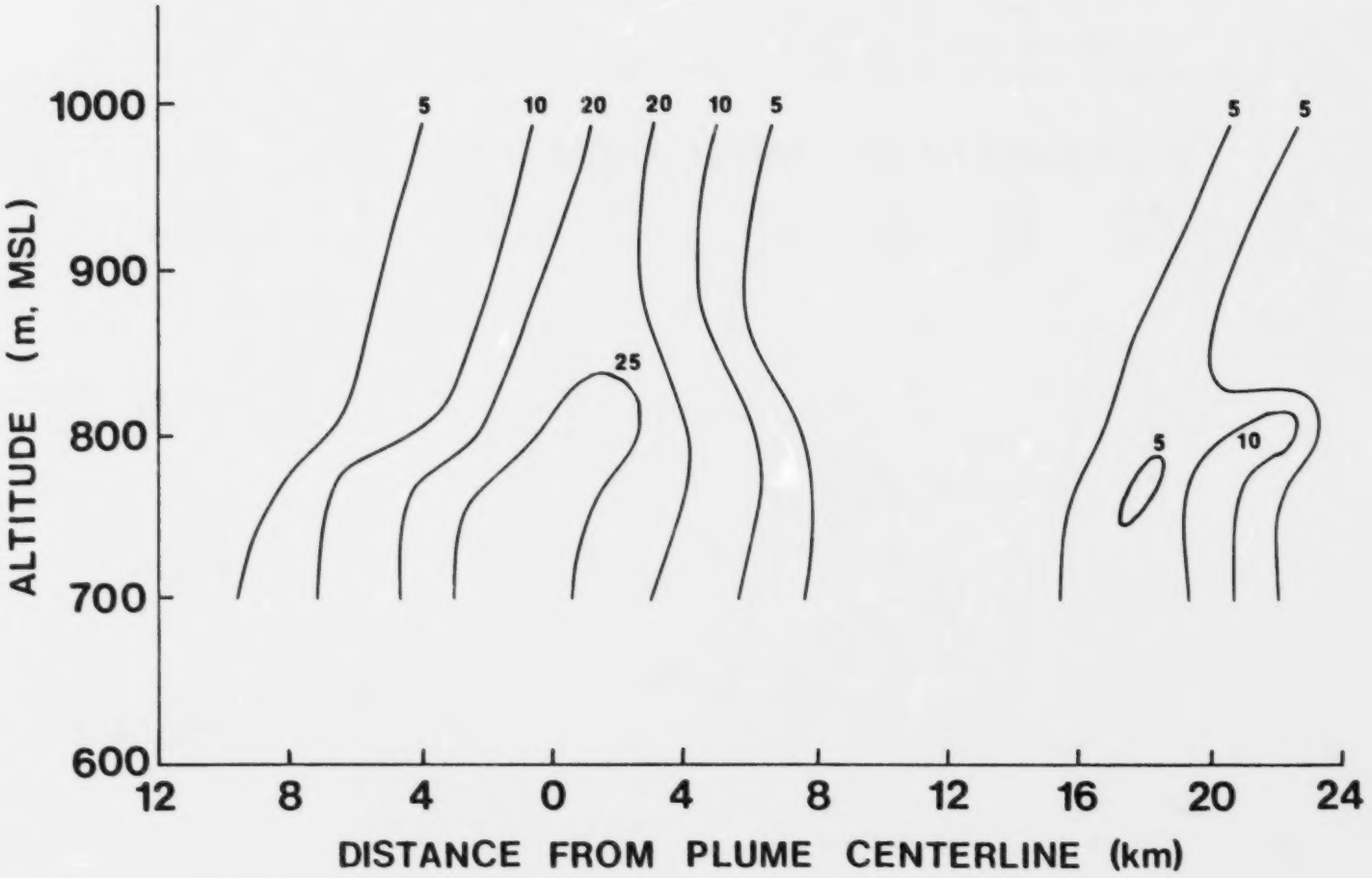

Figure 14. Contours of sulfur dioxide concentration (in ppb) measured perpendicular to the axis of the plume from the Widows Creek Power Plant on August 23, 1978 at 50 km downwind. The sulfur dioxide concentration in background air was between 1 and 4 ppb.

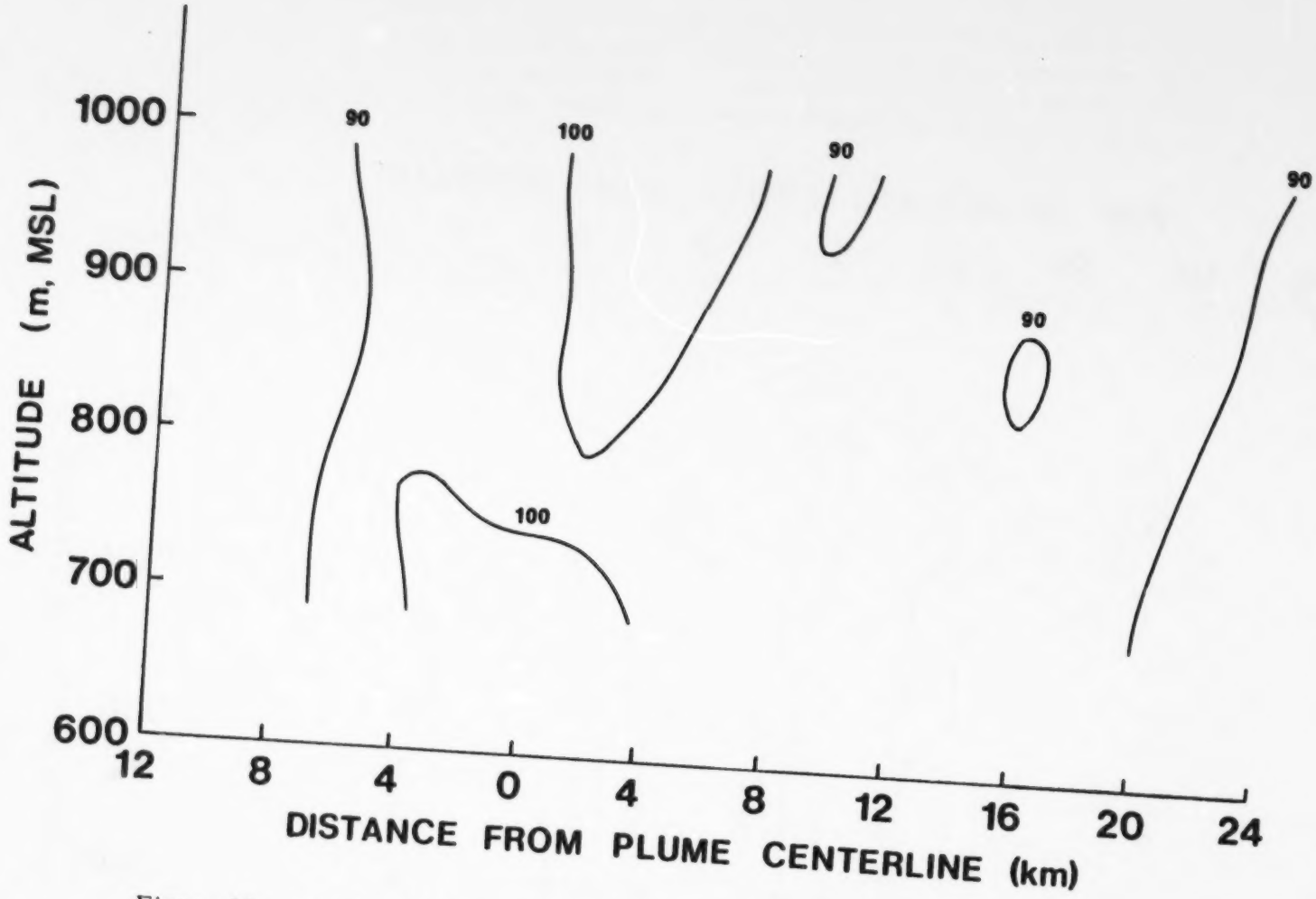

Figure 15. Contours of ozone concentration (in ppb) measured perpendicular to the axis of the plume from the Widows Creek Power Plant on August 23, 1978 at 50 km downwind.

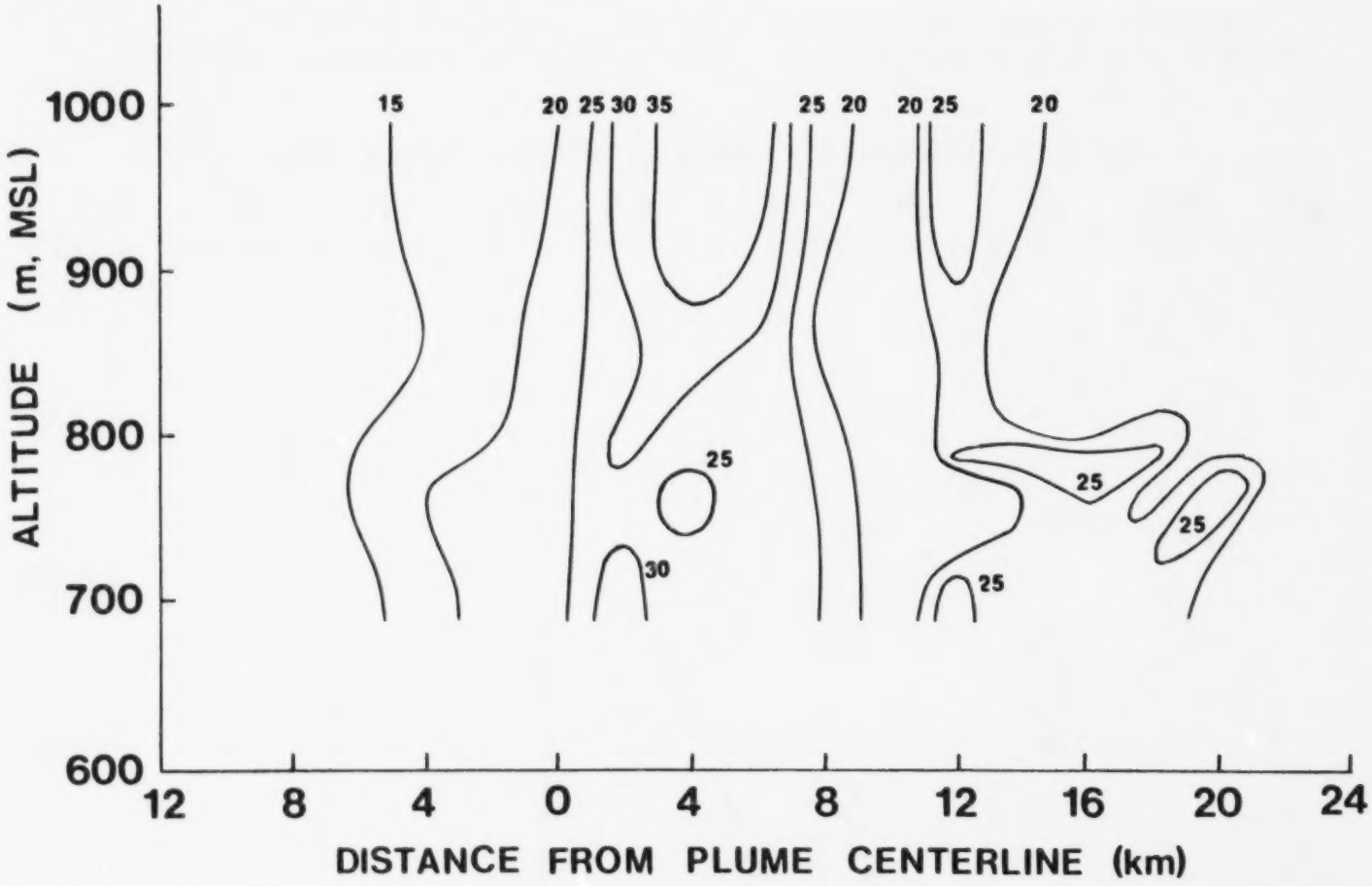

Figure 16. Contours of condensation nuclei concentration (in units of 10^3 cm^{-3}) measured perpendicular to the axis of the plume from the Widows Creek Power Plant on August 23, 1978 at 50 km downwind. The concentration of CN in the background air was between 8 x 10^3 and 15 x 10^3 cm^{-3}.

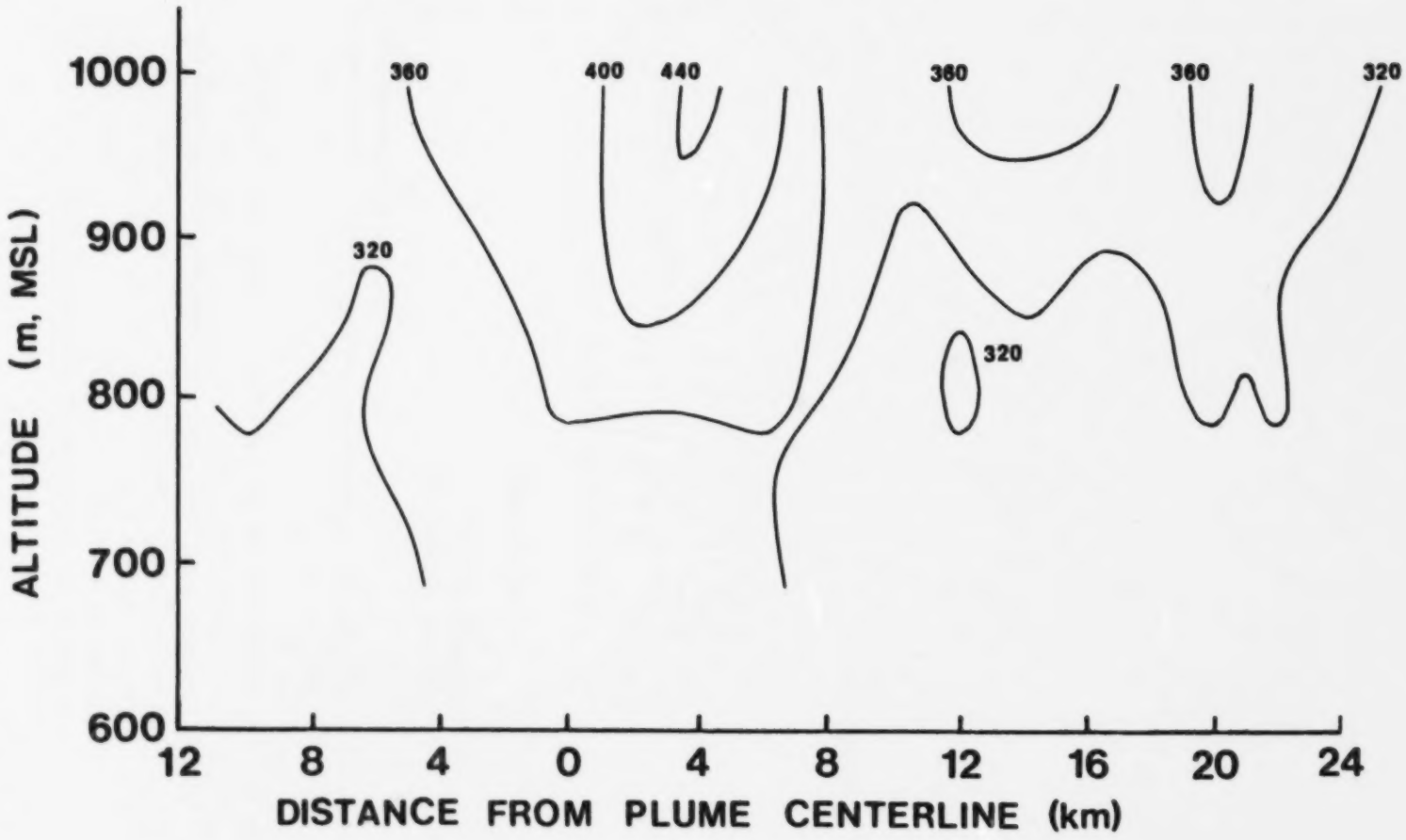

Figure 17. Contours of light scattering coefficient due to particles (in units of 10^{-6} m^{-1}) measured perpendicular to the axis of the plume from the Widows Creek Power Plant on August 23, 1978 at 50 km downwind.

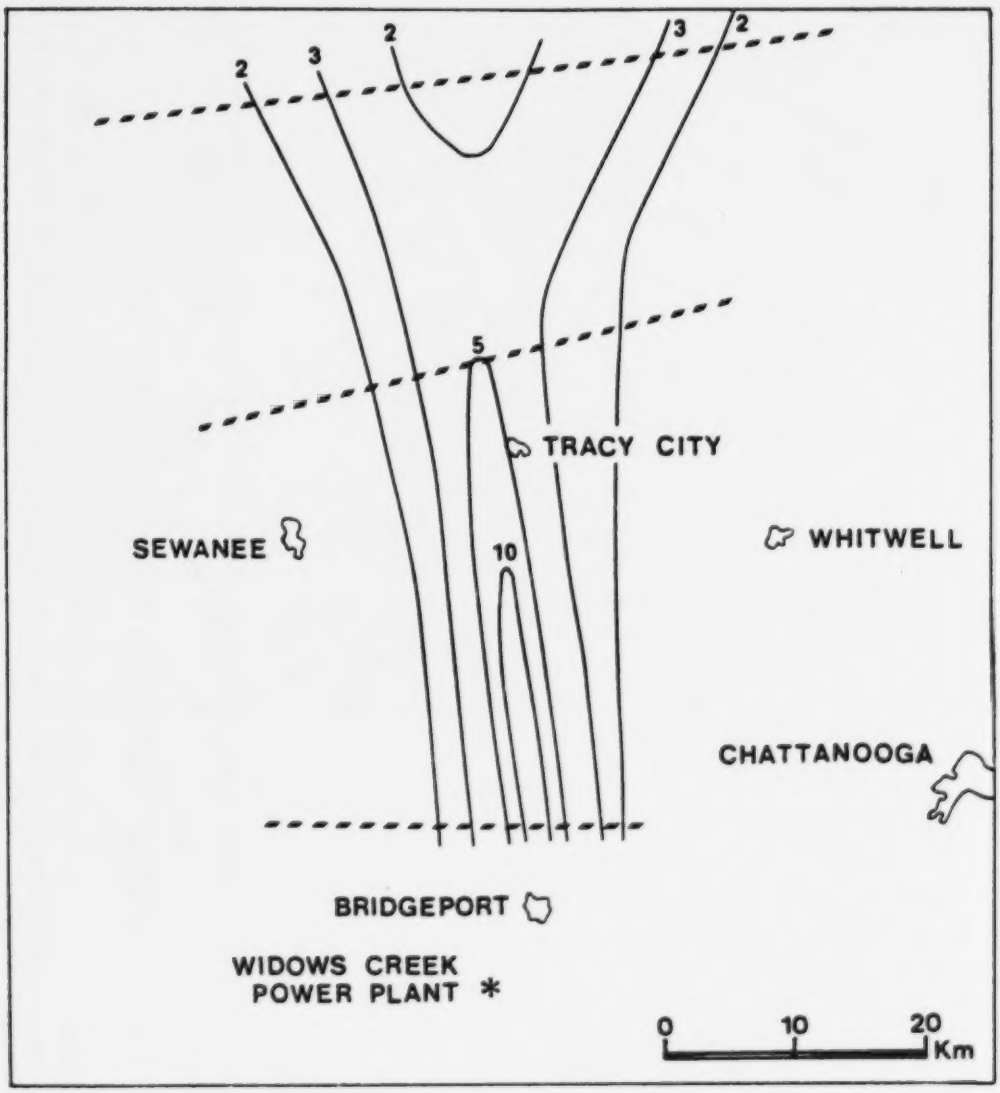

Figure 18. Contours of sulfur dioxide concentration (in ppb) measured parallel to the axis of the plume (looking down from above) from the Widows Creek Power Plant on August 23, 1978 at an altitude of 975 m (MSL). The sulfur dioxide concentration in background air was between 1-2 ppb. The flight paths of the aircraft are indicated by the dashed lines.

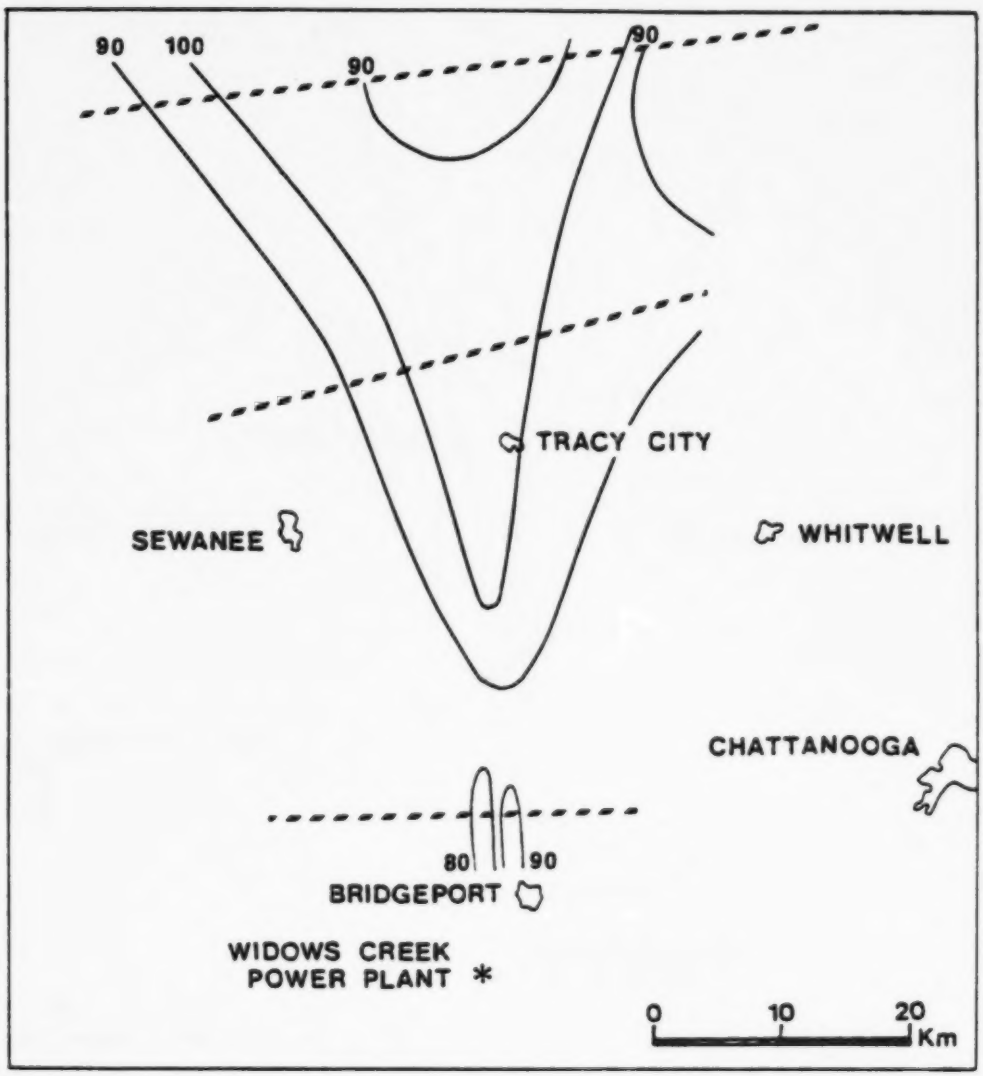

Figure 19. Contours of ozone concentration (in ppb) measured parallel to the axis of the plume (looking down from above) from the Widows Creek Power Plant on August 23, 1978 at an altitude of 975 m (MSL). The ozone concentration in background air was between 80 and 90 ppb. The flight paths of the aircraft are indicated by the dashed lines.

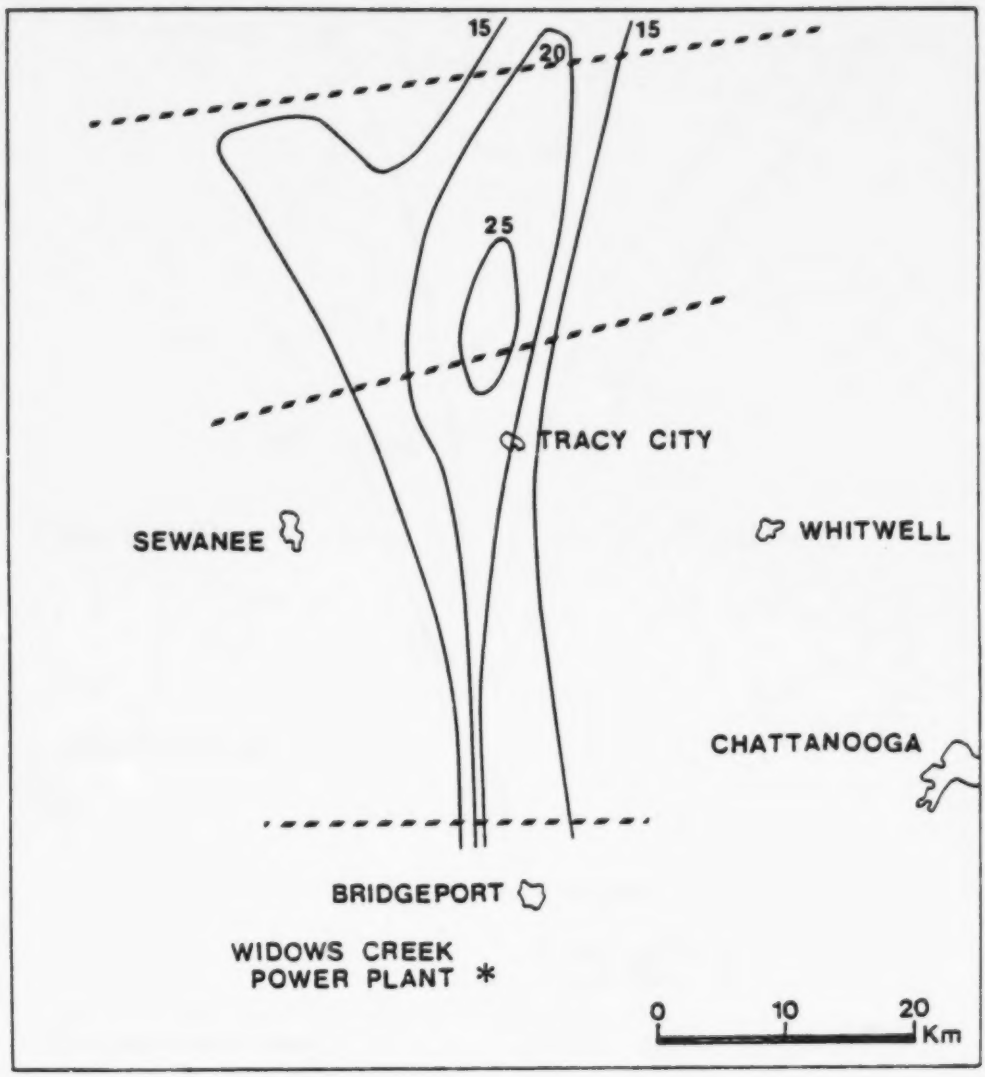

Figure 20. Contours of condensation nuclei concentration (in units of 10^3 cm^{-3}) measured parallel to the axis of the plume (looking down from above) from the Widows Creek Power Plant on August 23, 1978 at an altitude of 975 m (MSL). The concentration of CN in the background air was between 9×10^3 and 15×10^3 cm^{-3}. The flight paths of the aircraft are indicated by the dashed lines.

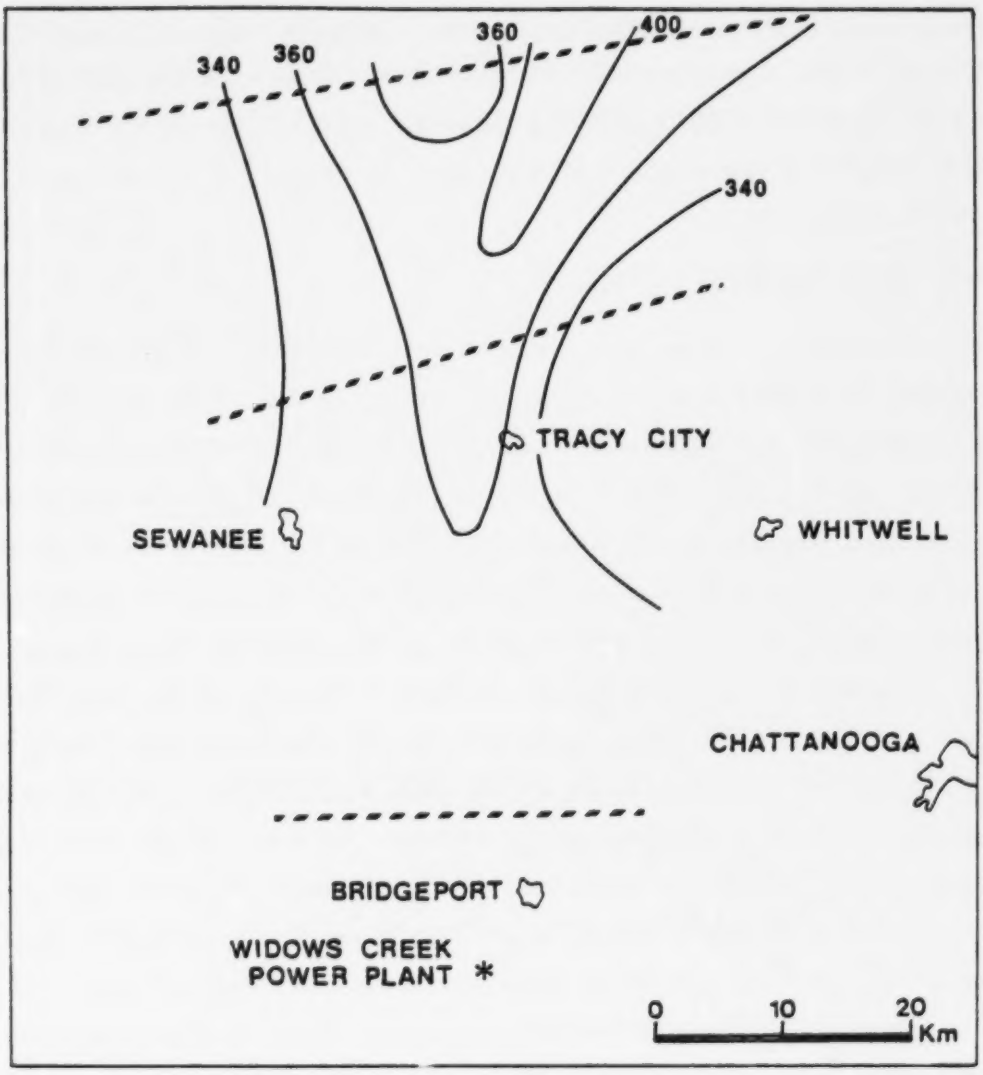

Figure 21. Contours of light scattering coefficient due to particles (in units of 10^{-4} m^{-1}) measured parallel to the axis of the plume (looking down from above) from the Widows Creek Power Plant on August 23, 1978 at an altitude of 975 m (MSL). The light scattering coeffieient due to particles, in the background air, increased downwind of the Widows Creek Plant; at 13 km downwind it was between 2.5×10^{-4} and 3.0×10^{-4} m^{-1} and at 50 -70 km downwind it was between 3.0×10^{-4} and 3.4×10^{-4} m^{-1}. The flight paths of the aircraft are indicated by the dashed lines.

increased downwind in both the background and in the plume; however, a maximum in the plume is evident beginning at the 50 - 70 km distance. The concentration of CN reached a maximum value at the 50 km distance, then decreased in the region which displays the higher levels of b_{sp} (Figures 20 and 21).

PARTICLE SIZE DISTRIBUTIONS

A summary of some of the size distribution results from the morning and afternoon flights are presented in Tables 7 and 8, respectively, along with the sulfate concentrations which were measured while the bagfills were obtained. As is seen in Table 7, during the morning flight the nuclei mode concentration was slightly higher in the plume at both distances downwind. The accumulation mode concentration was at about background levels at 11-14 km range, and higher than background levels at the 50 km range. These results confirm the increases in light scattering which were observed (Figures 13 and 17). Between these two ranges, the plume had diluted by about a factor of 4. Normalizing the average plume above ambient nuclei and accumulation mode volume concentrations, by the above ambient SO_2 concentrations, we find the ratio to be $\sim 4 \times 10^{-4}$ and $0.002 \mu m^3\ cm^{-3}\ ppb^{-1}$ for the nuclei mode and ~0 and $0.32\ \mu m^3\ cm^{-3}\ ppb^{-1}$ for the accumulation mode at the 11-14 km and 50 km distances downwind, respectively. Thus, both of these modes were increasing downwind. The secondary particle volume in the nuclei mode amounted to about 0.6% of the total secondary particle volume, at the 50 km distance.

During the afternoon flight, both the nuclei and accumulation mode concentrations were near background levels at the 13 km distance (Table 8), but were above background at the further distances. The ratio of the average plume above ambient nuclei and accumulation mode volume concentrations to the above ambient SO_2 concentrations are about 0.013 and 0.006 $\mu m^3\ cm^{-3}\ ppb^{-1}$ for the nuclei mode, and 2.0 and $2.3\ \mu m^3\ cm^{-3}\ ppb^{-1}$ for the accumulation mode at the 50 and 70 km distances, respectively.

TABLE 7. SUMMARY OF SIZE DISTRIBUTION MEASUREMENTS TAKEN ON THE MORNING OF AUGUST 23, 1978

Time	Nuclei Mode Volume Concentration ($\mu m^3\ cm^{-3}$)	Accumulation Mode Volume Concentration ($\mu m^3\ cm^{-3}$)	SO_2 Concentration (ppb)	Sulfate * Concentration ($\mu gS\ m^{-3}$)	Comments
09:58:17	0.051	24.	77.		762 m altitude
10:15:32	0.12	12.	100.		792 m altitude
10:24:54	0.15	17.	116.	1.8	853 m altitude — 11-14 km downwind
10:32:22	0.11	20.	90.		914 m altitude
10:41:06	0.065	17.	46.		975 m altitude
	0.099 , 0.041	18. , 4.4	86. , 27.	1.8	Average and standard deviation at 11-14 km
11:52:03	0.061	29.	22.		762 m altitude
12:05:49	0.12	29.	29.		701 m altitude
12:14:51	0.11	29.	21.	5.8	792 m altitude — 50 km downwind
12:28:22	0.14	25.	23.		853 m altitude
12:36:41	0.14	35.	20.		975 m altitude
	0.11 , 0.032	29. , 3.6	23. , 3.5	5.8	Average and standard deviation at 50 km
11:08:34	0.042	21.	0.		914 m altitude
11:15:12	0.088	23.	1.	4.4	914 m altitude — Background samples
11:19:26	0.054	22.	2.		914 m altitude
	0.061 , 0.024	22. , 1.0	~1.	4.4	Average and standard deviation of background samples

* Sulfate concentrations are from Two-Mass filters which were being exposed while the bag samples were obtained; often several bagfills were obtained while one filter was being exposed.

TABLE 8. SUMMARY OF SIZE DISTRIBUTION MEASUREMENTS TAKEN ON THE AFTERNOON OF AUGUST 23, 1978

Time	Nuclei Mode Volume Concentration ($\mu m^3 cm^{-3}$)	Accumulation Mode Volume Concentration ($\mu m^3 cm^{-3}$)	SO_2 Concentration (ppb)	Sulfate * Concentration ($\mu gS\ m^{-3}$)	Comments
14:50:30	0.12	23.	4.	6.7	853 m altitude } 70 km
15:04:17	0.057	22.	4.	6.6	792 m altitude } downwind
	0.088 , 0.044	22. , 0.71	4.	6.6 , 0.071	Average and standard deviation at 70 km
17:14:49	0.14	24.	4.	7.0	
17:23:10	0.11	24.	4.	} 7.1	} 975 m altitude
17:28:22	0.15	23.	7.		} 50 km downwind
17:32:54	0.11	24.	7.		
	0.13 , 0.021	24. , 0.50	5.5 , 1.7	7.0 , 0.071	Average and standard deviation at 50 km
18:15:14	0.087	15.	26.	5.6	
18:36:25	0.037	17.	13.	} 5.4	} 975 m altitude
18:41:06	0.063	15.	12.		} 13 km downwind
18:45:45	0.090	15.	39.		
	0.069 , 0.025	16. , 1.0	22. , 13.	5.5 , 0.14	Average and standard deviation at 13 km

* Sulfate concentrations are from Two-Mass filters which were being exposed while the bag samples were obtained; often several bagfills were obtained while one filter was being exposed.

TABLE 8 (continued)

Time	Nuclei Mode Volume Concentration ($\mu m^3 cm^{-3}$)	Accumulation Mode Volume Concentration ($\mu m^3 cm^{-3}$)	SO_2 Concentration (ppb)	Sulfate * Concentration ($\mu gS\ m^{-3}$)	Comments
17:48:09	0.060	15.	1.	4.5	975 m altitude Background samples
17:52:16	0.048	14.	1.		
17:56:14	0.061	14.	1.		
18:52:08	0.10	13.	0.	6.4	
18:54:59	0.082	17.	2.		
	0.070 , 0.021	15. , 1.5	1. , 0.71	5.4 , 1.3	Average and standard deviation of background samples

* Sulfate concentrations are from Two-Mass filters which were being exposed while the bag samples were obtained; often several bagfills were obtained while one filter was being exposed.

Since the concentrations were near background levels at the 13 km distance, both modes increased between 13 and 50 km. The secondary particle volume in the nuclei mode was about 0.6% of the total secondary particle volume at the 50 km distance and about 0.3% at the 70 km distance. As was the case on August 19, the nuclei mode decreased at the farthest range downwind (70 km).

Some examples of the particle number, surface, and volume distributions, for the afternoon flight, measured at the 13, 50, 70 km distances and in the background air are presented in Figures 22 and 23. The increase in the accumulation mode at 50 and 70 km is evident in Figures 21 and 23. The relative depression of the portion of the number distribution less than 0.05 μm, at the 70 km range (Figure 23), is evident, in agreement with the lower concentration of condensation nuclei observed at this range (Figure 20). As was the case on August 17, these emissions are not a significant source of coarse (> 1.0 μm) particles.

GAS-TO-PARTICLE CONVERSION

The sulfate levels measured in both the plume and the background air were among the highest observed during the entire SCRUB project. These measurements (see Tables 7 and 8) indicate that the sulfate concentrations in the plume were increasing downwind during the morning and afternoon flights. However, the concentration in the background air was also increasing during this sampling period. Consequently, the amount of sulfate produced by the plume is uncertain.

Most of the sulfate filter samples which were collected during the morning flight were collected by activating the Two-Mass sampler at the beginning of the aircraft traverse and then shutting it off at the end of the traverse. This was repeated, using the same filter sample, until the set of traverses at that distance downwind were completed. For these traverses, the aircraft was generally in the plume for less than half of the time the

filter was being exposed. Consequently, the sulfate samples obtained during the plume traverses at 11-14 km and 50 km downwind are probably highly diluted by background air.

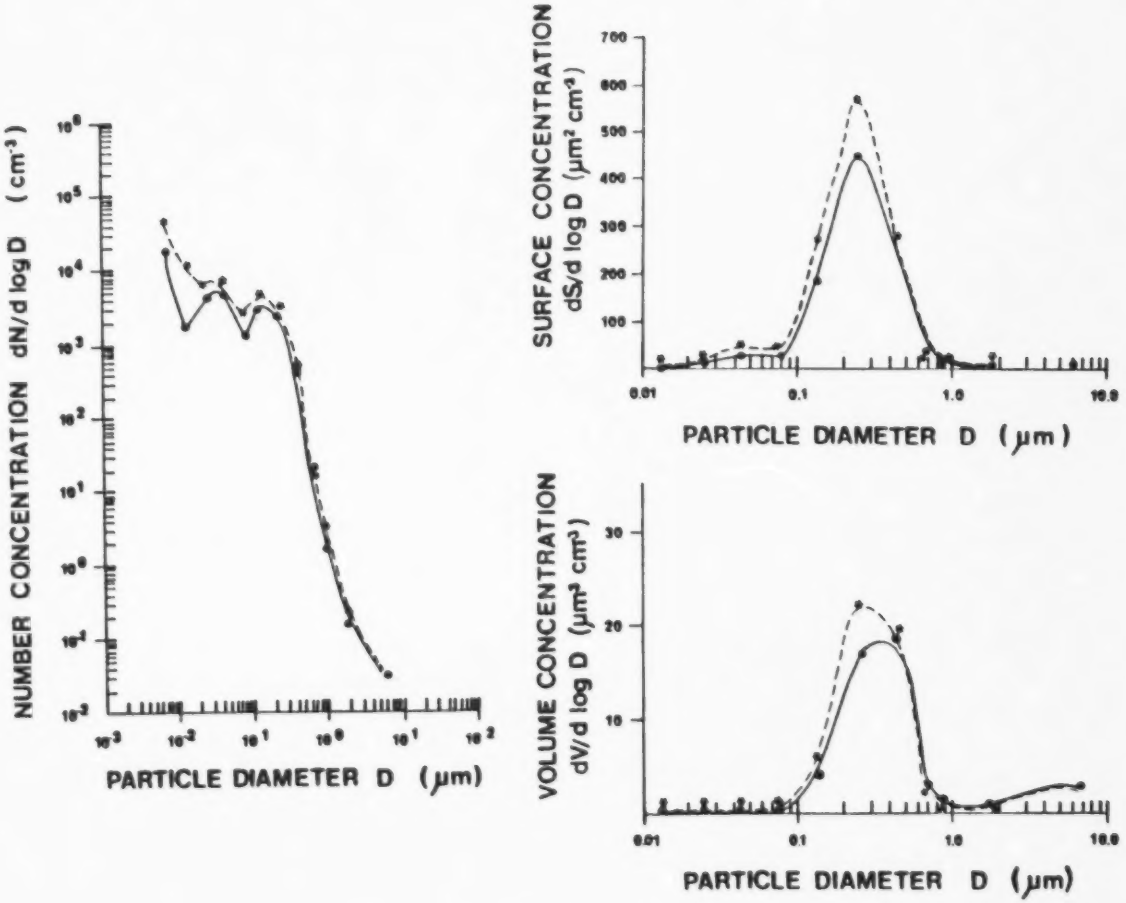
NUMBER CONCENTRATION dN/d log D (cm⁻³)
PARTICLE DIAMETER D (μm)
SURFACE CONCENTRATION dS/d log D (μm² cm⁻³)
PARTICLE DIAMETER D (μm)
VOLUME CONCENTRATION dV/d log D (μm³ cm⁻³)
PARTICLE DIAMETER D (μm)

Figure 22. Particle number (dN/dlog D), surface (dS/dlog D), and volume (dV/dlog D) distributions, measured in the plume of the Widows Creek Power Plant on August 23, 1978 at 13 km downwind (dotted line) and at an altitude of 975 m (MSL). A distribution taken in the background air at an altitude of 975 m is also presented (solid line).

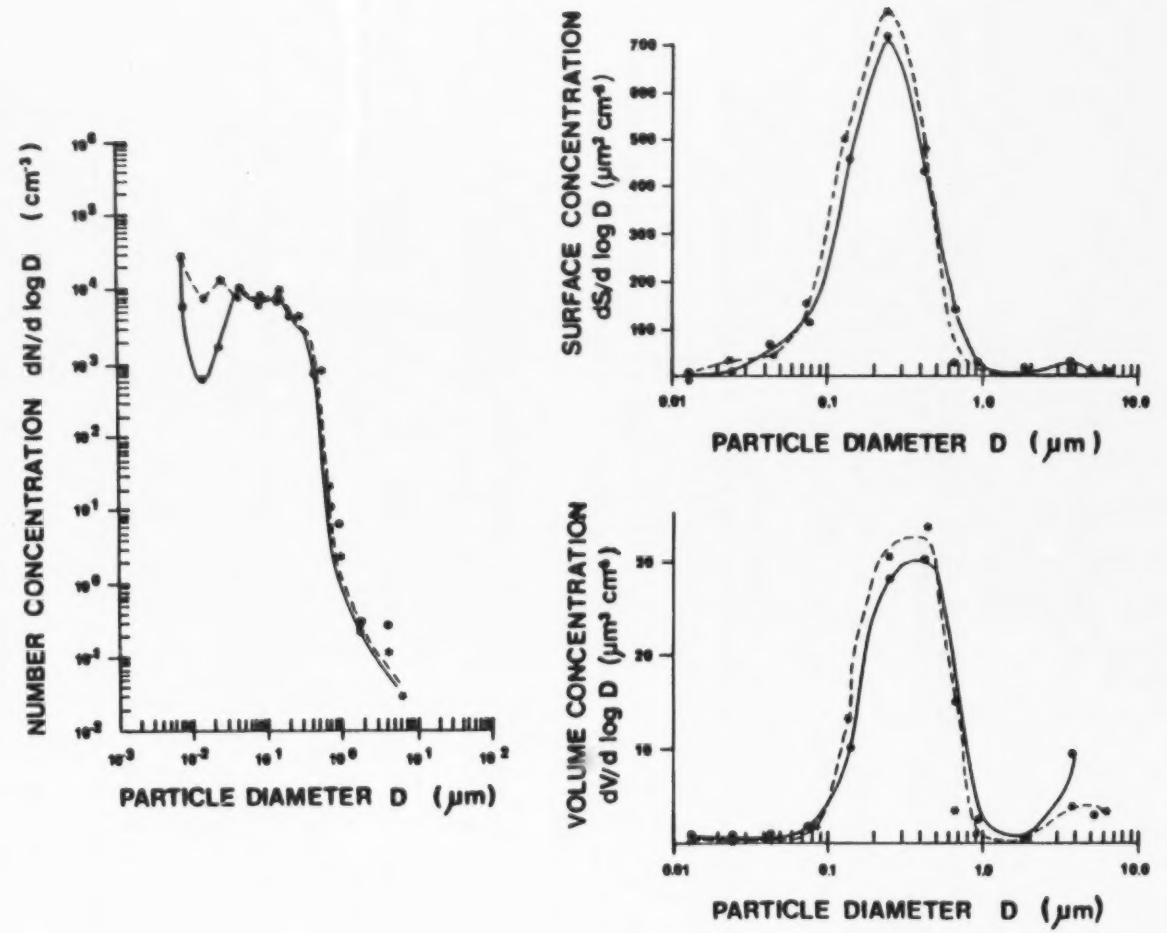
NUMBER CONCENTRATION dN/d log D (cm⁻³)
PARTICLE DIAMETER D (μm)
SURFACE CONCENTRATION dS/d log D (μm² cm⁻³)
PARTICLE DIAMETER D (μm)
VOLUME CONCENTRATION dV/d log D (μm³ cm⁻³)
PARTICLE DIAMETER D (μm)

Figure 23. Particle number (dN/dlog D), surface (dS/dlog D), and volume (dV/dlog D) distributions, measured in the plume of the Widows Creek Power Plant on August 23, 1978 at 50 km downwind and an altitude of 975 m (MSL) (dotted line), and at 70 km downwind at an altitude of 853 m (solid line).

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APPENDIX A

GAS-TO-PARTICLE CONVERSION RATE UNCERTAINTIES

In this appendix the uncertainties in the rate of gas-to-particle conversion are estimated for the August 17 case study. If the uncertainty in each of the measurements of the sulfate function (i.e. the ratio of the above ambient sulfate to the above ambient total sulfur) is given by the standard deviation of the values, σ, then the uncertainty in the mean value, σ_m, is given by* $\sigma/\sqrt{N}$, where N is the number of measurements. N = 2 for the measurements made at 3 and 9 km downwind on August 17. The standard deviations of the sulfate fraction (see Section 4) are 0.45% and 0.28% for the 3 and 9 km distances, respectively. Thus the uncertainties in the mean sulfate function at 3 and 9 km are 0.32 and 0.20%, respectively. The rate of gas-to-particle conversion, R, between the two distances is

$$R = \frac{1}{T} \ (f_9 - f_3), \qquad \text{A-1}$$

where T is the travel time between the two distances, f is the mean sulfate fraction and the subscript refers to the distance downwind. The uncertainty in R is given by *

$$\sigma_R^2 = \sigma_{m9}^2 \left(\frac{\partial R}{\partial f_9}\right)^2 + \sigma_{m3}^2 \left(\frac{\partial R}{\partial f_3}\right)^2 + \sigma_T^2 \left(\frac{\partial R}{\partial T}\right)^2 \qquad \text{A-2}$$

where σ_T is the uncertainty in the plume travel time which is estimated to be about 20% of T (T = .55 hr) or .11 hr. Then, from A-1 and A-2,

$$\sigma_R^2 = \sigma_{m9}^2 T^{-2} + \sigma_{m3}^2 T^{-2} + \sigma_T^2 \left[-(f_9 - f_3)T^{-2}\right]^2 \qquad \text{A-3}$$

Substituting in values (f_9 = 1.4%, f_3 = 0.68%, see Section 4) we find σ_R = 0.75%.

* For the derivation of these formulas see, for example, Bevington (1969).

σ_R provides a measure of the errors in the conversion rate associated with the random sampling errors. These errors are about 56% of the conversion rate. As was noted in Section 3 there may be additional systematic errors associated with the measurement technique which suggest that the sulfate concentrations may be low by about a factor of 2. The conversion rate would be low by the same factor (see equation A-1).